AN INVESTIGATION OF THE FACTORS AFFECTING MERCURY ACCUMULATION IN LAKE TROUT, *SALVELINUS NAMAYCUSH*, IN NORTHERN CANADA

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By

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ABSTRACT

The major aim of this thesis project was to determine the variables that most explain the elevated mercury concentrations in lake trout (*Salvelinus namaycush)*, a predatory aquatic fish species in some lakes in northwestern Canada. High mercury concentrations in lake trout in other regions have been associated with the biological features of the fish and various chemical and physical aspects of their aquatic ecosystems. Data including lake trout age, length, weight, and stable isotope values, water chemistry, latitude, and lake and watershed area were collected, compiled and then included in statistical analyses of the factors affecting mercury concentration in the muscle of lake trout from a series of lakes from the Mackenzie River Basin (MRB) in the Northwest Territories (NT), Canada. These results are reported in Chapter 2. Fish age and lake surface area were the most important variables affecting mercury concentrations. However mercury concentration in muscle also was significantly ($p < 0.05$) related to: fish length, weight, and $\delta^{13}C$; watershed area to lake area ratio; and to total mercury concentration in zooplankton and water. These variables were run through best subsets analyses and multiple regressions in order to determine the regression equation most efficiently capable of predicting mercury concentration in lake trout in unstudied lakes in the MRB region. The resulting equation was:

$$\log Hg = 0.698 - (0.0156 \times latitude) + (0.0031 \times age) + (0.000535 \times length) - (0.245 \times \log lake\ area) + (0.00675 \times watershed\ area/lake\ area\ ratio),\ r^2 = 0.73$$

Small lakes located in the southern NT and dominated by large and/or old lake trout are most likely to have lake trout whose mean mercury concentrations exceed 0.5 μg/g; the guideline for the commercial sale of fish. Latitude may be linked to mean annual temperature (and variables such as duration of ice cover, summer water temperature) while fish age and length may be related in part to fishing pressures and growth rates on these lake populations.

In chapter 3, a more in-depth study was undertaken to investigate of role of feeding and relative tropic level in the bioaccumulation of mercury in lake trout. This was accomplished by comparing MRB lake trout population characteristics with those from a series of lakes in northern Alberta and Saskatchewan (NAS). The

two population groups were compared with respect to size, age, growth rates, and mercury concentrations. In addition, trophic and mercury biomagnification relationships, as inferred from stable carbon and nitrogen isotope analyses, for the two lake trout populations were compared. Lake trout from the NT exhibited significantly higher mercury concentrations than those from the NAS lakes ($p < 0.001$). Mercury concentrations in biota (including lake trout, forage fish, benthic invertebrates and zooplankton) were positively and significantly correlated to $\delta^{15}N$ values in all lakes in both of the study areas ($p < 0.001$). Mercury biomagnification in the NT lakes, as estimated from the slope of $\delta^{15}N$ versus mercury concentration, was lower than in the NAS lakes. Thus, mercury biomagnifies more slowly in NT lake trout, but because of their greater mean age, reaches higher values than in NAS lakes. Northwest Territory lake trout generally exhibited more negative $\delta^{13}C$ values, indicating more pelagic feeding habits than in NAS lakes: higher mercury concentrations previously have been associated with more pelagic feeding.

Finally, the relationship between mercury levels and growth rates in lake trout was investigated by comparing NAS and NT lake trout populations. These results are reported in chapter 4. Lake trout from the NT lakes grew at a slower rate (10.4 mm per year) than those from the NAS lakes (35.1 mm per year). Log mercury concentration was inversely correlated ($p < 0.001$) with growth rate for both lake trout populations; however, growth rate explained more of the variation in mercury level in the NT lakes than in the NAS lakes (NT, $r^2 = 0.11$, $p < 0.001$; NAS, $r^2 = 0.03$, $p = 0.024$). However, the correlation between mercury concentration and growth rate in the NAS study area improved when Reindeer Lake, possibly affected by anthropogenic inputs, was removed from the analyses ($r = 0.13$, $p = 0.001$). Therefore, lower mercury levels in lake trout are associated with higher growth rates through growth dilution. The higher mercury concentrations in NT lake trout are due not only to the old age of the fish, but to slower growth rates as well.

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LIST OF ABBREVIATIONS

ANOVA	Analysis of variance
ATSDR	Agency for Toxic Substances and Disease Registry
BRCL	Bromine chloride
$\delta^{13}C$	Carbon^{13} stable isotope
CF-IRMS	Confinuous flow isotope ratio mass spectrometer
CFIA	Canadian Food Inspection Agency
Chl*a*	Chlorophyll *a*
CSFG	Commercial sale of fish guideline
DFO	Fisheries and Oceans Canada
DO	Dissolved oxygen
DOC	Dissolved organic carbon
dw	Dry weight
ESRI	Environmental Systems Research Institute
FCFG	Frequent consumer of fish guideline
FWI	Freshwater Institute
FWMF	Food web magnification factor
GC	Gas Chromatograph
GIS	Geographic information systems
GRRs	Growth rate residuals
HCl	Hydrochloric acid
Hg	Mercury
MCRs	Mercury concentration residuals
MeHg	Methylmercury
MRB	Mackenzie River Basin
MRBB	Mackenzie River Basin Board
N	Number of sampled units
$\delta^{15}N$	Nitrogen^{15} stable isotope
NAS	Northern Alberta and Saskatchewan
NC	No concern
NCP	Northern Contaminants Program
NEI	Northern Ecosystem Initiative
NIST	National Institute of Standards and Technology
NRC	National Research Council
NT	Northwest Territories
NTDB	National Topographic Database
NWRI	National Water Research Institute
OH^-	Hydroxide ion
PANP	Prince Albert National Park
POPs	Persistent organic pollutants
RMWI	Recommended maximum weekly intake
SK	Saskatchewan
SRB	Sulphur reducing bacteria
SRRB	Sahtu Renewable Resources Board
THg	Total mercury
TP	Total phosphorous
TSRI	Toxic Substance Research Initiative
UNEP	United Nations Environmental Programme
US EPA	US Environmental Protection Agency
WHO	World Health Organization
ww	Wet weight

1.0 GENERAL INTRODUCTION

1.1. Historic and current uses of mercury

Mercury is an inorganic, naturally occurring element with ubiquitous distribution throughout the global environment. The Chinese, Hindu, Greek, and Roman people have recognized its presence and applications since ancient times. Egyptians used mercury to form amalgams in 1500 BC, Greeks used it in ointments, and Romans used it in cosmetics (D'Itri 1977). It has also been used in the extraction of gold because of its ability to combine with other metals and form amalgams. In addition to being used widely in the past, mercury has many current applications with approximately 10,000 tonnes of mercury mined per year to supply global requirements (UNEP 2002). Current uses include its incorporation in thermometers, barometers, batteries, other scientific apparatus', streetlights, fluorescent lamps and advertising signs (US EPA 1997).

As long as people have been using mercury, they have known about its toxic properties (UNEP 2002). The first recorded case of mercury poisoning occurred as early as 50 BC when the Greeks and Romans were using mercury as a component in ointments and cosmetics. Today we recognize mercury as a potentially dangerous neurotoxin that can result in death under certain circumstances (USA ATSDR 2001). Mercury can be taken up via inhalation and consumption. The greater uptake routes is through the consumption of foods which contain mercury. Many countries regulate mercury intake though commercial sale and consumption advisories. For example, the Canadian Food Inspection Agency's (CFIA) guideline for the commercial sale of fish (CSFG), both freshwater and marine, is 0.5 μg Hg/g wet weight (ww) in muscle tissue and Health Canada has established a guideline of 0.2 μg Hg/g ww for frequent consumers of fish (FCFG) (CFIA 2002; Shilts and Coker 1995; Stephens 1995). The Northwest Territories (NT) issued consumption advisories for many lakes in the NT based on mercury concentrations in fish tissues; many 'First Nations' people in the region rely heavily on fish as an important

component of their traditional diet and thus fall into the high-risk category of frequent consumers (Chan *et al.* 2003).

1.2. Sources and forms of environmental mercury

Mercury is an element naturally present in the earth's crust. It occurs as a liquid at room temperature, and evaporates easily into the atmosphere. Mercury is emitted to the environment naturally through volcanic eruptions and seismic activity, the weathering of rocks and soils, photo-reduction of divalent mercury in natural waters and biological formation from methylation of elemental or dimethylmercury (UNEP 2002). Increasing amounts are also released through human activity; since industrialization the amount of mercury found in the environment has increased by a factor of three (Porcella *et al.* 1996). Mercury is released through many industrial processes including energy production (coal-fired power plants), base metal smelting, gold mining, and waste incineration (Neimi 1998; USA ATSDR 2001). Therefore mercury pollution levels are highest in industrial areas where these processes are occurring. In 1995, Canada's largest sources of mercury emissions were the nonferrous (primary base-metals) industry, coal-fired power plants, and municipal solid waste incinerators (Neimi 1998). This released mercury is transported through the air, where it can travel long distances, and is eventually deposited on water and land either as dry particles, or through wet precipitation (UNEP 2002). It is also transported over the earth's surface through effluent discharge into flowing rivers and via ocean currents. High environmental mercury concentrations also occur in areas affected by reservoir creation. Experimental studies have shown that reservoir creation can increase the total mercury (THg) and methylmercury (MeHg) yields by 40-fold (Montgomery *et al.* 2000; St. Louis *et al.* 2004).

Mercury has three valence states (Hg^0, Hg^{1+}, and Hg^{2+}) and occurs in the environment in its metallic form as well as in various inorganic and organic complexes. The natural mercury biogeochemical cycle involves degassing of mercury from soils and surface waters, atmospheric transport, deposition of mercury back to land and surface water, sorption of mercury onto soil or sediment particles, and re-release back to the water column and atmosphere (see Figure 1.1) (NRC 2000). Sediment cores collected from central and northern Canada show an

increase in mercury deposition over time, possibly due to increased anthropogenic release of mercury to the atmosphere (Lockhart *et al.* 1998).

This complex cycle, including mixing of mercury from various sources, makes it difficult to trace mercury in a given region back to its original source (USA ATSDR 2001). Different forms and oxidation states of mercury are interchangeable in atmospheric, aquatic, and terrestrial environments, and the proportions of the different mercury species depend on the combined effects of numerous physicochemical and biological variables (Jackson 1997).

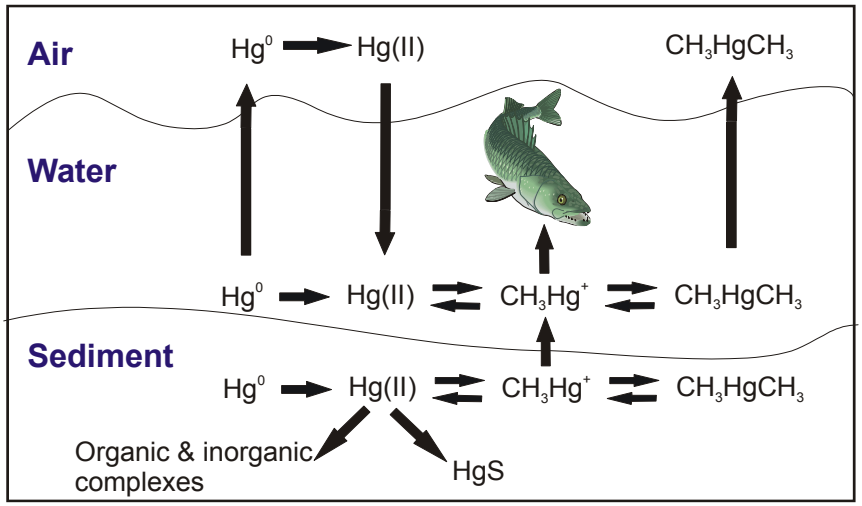


Figure 1.1. Mercury transformations within the environment (adapted from Winfrey and Rudd 1990).

1.3. Exposure and adverse health effects

The form of mercury of most concern from a human and ecosystem health perspective is MeHg. Methylmercury is a soluble, stable, and persistent compound with a high affinity for organic matter. This affinity coupled with the stability and persistence of MeHg result in its biomagnification in food webs, leading to high body burdens at the top of these food webs, i.e., predaceous fish, birds, and mammals, including humans. Fish consumption is the main source of human exposure to MeHg (NRC 2000), with the exception of the coastal Arctic communities who are exposed mainly through the consumption of marine mammals.

1.3.1. Human health effects

Human exposure to MeHg through the consumption of freshwater and marine fish is the dominant health concern with mercury in the environment (Fitzgerald *et al*. 1998). Methylmercury is readily absorbed from the gastrointestinal tract, enters the blood, and is well distributed to all organs including the brain. It is also able to cross the placenta and enter the fetal brain. Once in the brain it is slowly converted to inorganic mercury. Individual exposure can be measured through hair (maternal), blood, and umbilical cord blood. High MeHg body burdens (3 μg Hg/kg) can lead to toxic manifestations of many forms (WHO 1976). The most common toxic effects are neurobehavioral, and the most sensitive populations are young children and developing fetuses. Consumption of MeHg can lead to toxic effects when the exposure is an acute high dose (30 to 50 mg Hg/ 70 kg person, WHO 1976), or when there is a chronic, lower dose exposure. A widely known example of chronic toxicity associated with MeHg is the now classic case of Minamata disease. Beginning in the early 1950s, Minamata Bay (Japan) was polluted by local industries producing acetaldehyde which used mercury as a catalyst, a practice now regulated by environmental regulations (Tamashiro and Arakaki 1985). Mercury released in various effluents into the bay was biomagnified in the food web reaching concentrations as high as 1.74 μg/g in the fish. Over 1953 to 1960, people were poisoned through consumption of these fish and displayed the classic signs of mercury poisoning, now called Minamata disease (Fujiki and Tajima 1992). This event is of historical importance because the cases of organic mercury poisoning in Minamata were the first known cases of poisoning through food chain transfer of environmental pollutants (Harada 1995). Symptoms displayed by patients included sensory disturbance and constriction of the visual field, coordination disturbance, dysarthria, hearing disturbance, tremor, and walking disturbance; symptoms varied from mild to serious depending on the patient (Tokuomi 1960). In 1962, it was determined that unborn children were also affected, and this condition was named ‘congenital Minamata disease’. These children displayed mental retardation, primitive reflexes, coordination disturbance, dysarthria, limb deformation, growth disorder, chorea-athetose, and hypersalivation (Harada 1986). Umbilical cord blood from these children was found to contain MeHg concentrations of approximately 1

μg Hg/g (Harada 1995). Once the cause of the disease was identified, remedial actions were put in place.

In Iraq in the early 1970s many people consumed flour made from grains treated with a fungicide containing MeHg (Bakir *et al.* 1973). These grains were meant for agricultural cultivation, not consumption. The wheat flour contained MeHg concentrations averaging 9.1 μg Hg/g. Many people, especially children exposed in utero, presented with symptoms consistent with those of MeHg toxicity (NRC 2000). Children presented with severe sensory impairments including deafness and blindness, general paralysis, hyperactive reflexes, cerebral palsy, and impaired mental development (Bakir *et al.* 1973).

Today few people outside third world and developing countries are exposed to high concentrations of mercury through localized anthropogenic sources. As improved environmental regulations are developed and enforced, these exposures will be reduced. While this is generally encouraging, recent investigations show that more subtle toxic effects can arise from low-level mercury concentrations. The US EPA states 5.8 μg Hg/L in maternal blood as the reference dose above which there is the danger of developing toxic manifestations including delayed development and cognitive changes in children (US EPA 1997). Health Canada has set 0.2 μg Hg/g as a FCFG; in the NT, consumption advisories have been issued for various lakes where fish exceed this guideline (Table 1.1). Different guidelines have been established for women of children-bearing years and children, given the greater vulnerability of the fetus and young children, than adult men.

1.3.2. Aquatic biota health effects

Methylmercury is effectively taken up by biota, and bioconcentration factors in the order of 10^4 to 10^7 have been reported (Stein *et al.* 1996; Ullrich *et al.* 2001). The high rate of bioconcentration is due to the high stability and lipid solubility of the MeHg molecule, its tendency to bind to sulfhydryl groups, and its slow rate of elimination. Methylmercury in fish tissues is acquired predominantly though dietary uptake with lesser amounts accumulated directly from water (Bodaly *et al.* 1993). In fish, mercury is quickly absorbed from the gut and attaches to red blood cells. As such, it is quickly transported to all organs. The route of uptake, whether from respiration through the gills, or by food ingestion, has little bearing on distribution through the body. Redistribution continues and ultimately skeletal muscle is the

main site of MeHg accumulation, where it binds to sulfhydryl groups in the protein. However, lower concentrations can be found in other fish tissues, including the spleen, kidney, and liver (Foster *et al.* 2000). Mercury concentrations can be quite high in the liver and kidney of marine mammals such as seals (Fisk *et al.* 2001).

The primary site of mercury toxicity in fish is the central nervous system. Long-term dietary exposure can lead to poor coordination, inability to feed, and diminished responsiveness (Scherer *et al.* 1975; Weiner and Spry 1996). In Minamata Bay, Japan, mercury contaminated fish were sluggish, exhibited abnormal movements, were emaciated and had brain lesions (Takeuchi 1968).

Fish-eating wildlife including loons, bald eagles, osprey, mink and otter can be exposed to high levels of MeHg in aquatic systems where mercury reaches high concentrations in these dietary items. The dose of MeHg required to induce neurological impairments is higher than that typically found in the environment, however, subtle behavioural alterations and reproductive impairment may be seen with chronic, low dietary exposure (Chan *et al.* 2003). Egrets from the Florida Everglades exposed to concentrations of 0.5 mg Hg/kg displayed behavioural alterations including decreased hunting frequency, shade-seeking, and a general decrease in activity (Bouton *et al.* 1999). Blood concentrations of 0.5 to 1.0 mg Hg/kg can lead to reproductive impairment including decreased hatchability and chick survival in many fish-eating bird species (Burger and Gochfield 1997).

Acute and/or chronic exposure to MeHg can affect survival and reproduction in fish-eating mammals including mink and otter, which are exposed to the highest concentrations as a result of biomagnification (Chan *et al.* 2003). Effects of acute and chronic MeHg toxicity include anorexia leading to weight loss, neural necrosis leading to impairment of sensory and motor skills, and ultimately death (Chan *et al.* 2003). Dietary concentrations of 1 mg Hg/Kg body weight are sufficient to cause neurotoxicity and death in adult mink and otter (Dansereau *et al.* 1999).

1.4. Mercury and methylmercury inputs to freshwater environments

Two basic pathways exist for mercury input into lakes: via runoff from the immediate drainage area, both surface and groundwater, and through direct wet precipitation and dry deposition on the lake surface (Rudd 1995; Iverfeldt and Johansson 1988). Watersheds can be a significant source of mercury to a lake

Table 1.1. Fish consumption advisories for various lakes in the Northwest Territories, Canada (MacKinnon, Northwest Territories Health and Social Services, *pers. comm.*, 20/02/2004*)*.

Lake	Species	RMWI (g/week) Adults	RMWI (g/week) Women	RMWI (g/week) Children
Aubry	Lake Whitefish[1]	NC	NC	NC
	Burbot[2]	NC	NC	NC
	Lake Trout[3]	NC	NC	155
Belot	Burbot	NC	NC	285
	Lake Trout	NC	NC	183
Colville	Lake Whitefish	NC	NC	NC
	Burbot	NC	NC	NC
	Lake Trout	NC	NC	200
	Northern Pike[4]	NC	NC	150
Cli	Lake Whitefish	NC	NC	NC
	Lake Trout	225	100	45
Little Doctor	Lake Whitefish	NC	NC	NC
	Lake Trout	NC	NC	NC
	Northern Pike	250	110	50
	Walleye[5]	250	110	50
Turton	Lake Whitefish	NC	NC	NC
	Lake Trout	200	140	85
Manuel	Lake Whitefish	NC	NC	NC
	Burbot	NC	NC	NC
	Northern Pike	430	185	85

[1] *Coregonus clupeaformis*
[2] *Lota lota*
[3] *Salvelinus namaycush*
[4] *Esox lucius*
[5] *Sander vitreus vitreus*
NC – do not pose a concern
RMWI – recommended maximum weekly intake

depending on the relative size of the watershed-to-lake area (Iverfeldt and Johansson 1998). Furthermore, there are three important sources of MeHg to aquatic systems: precipitation, runoff from wetlands and in-lake methylation (Rudd 1995). For drainage lakes where atmospheric input is low (low precipitation, non-polluted areas), Rudd (1995) suggested that both the watershed and internal production are important sources of MeHg.

Mercury in the environment is now studied in areas distant from direct human impact, including Arctic Canada and Greenland. Though mercury has always been present at background levels, there is a long-term trend of increasing mercury in sediments in many remote areas due to atmospheric deposition. Releases from metal mining and industrialization activities are possible explanations for this trend (Bindler *et al.* 2001). Increasing mercury concentrations in these relatively pristine remote environments are due in part to increased atmospheric inputs of elemental mercury (Lockhart *et al.* 1998). This was determined through sediment core studies which investigated mercury deposition over time in lakes throughout northern and central Canada, and in Hudson Bay. These cores show that mercury fluxes have increased two-fold in the last half century (Lockhart *et al.* 1998). This enrichment is thought to be anthropogenic in origin, with the greatest enrichments occurring in central and southern Canada closer to industrial sources (Lockhart *et al.* 1998). Similarly ice coring studies conducted in Wyoming, USA found that over the last 100 years anthropogenic sources contributed 70% of the THg input (Schuster *et al.* 2002).

1.5. Mercury methylation and demethylation transformations

The rate of mercury methylation is greater in aquatic sediments than in the water column (Ullrich *et al.* 2001). However, more MeHg may ultimately be produced in the water column because of its larger volume relative to the thin layer of biologically active surficial sediments (Ullrich *et al.* 2001). Methylation occurs through both biotic and abiotic processes; abiotic methylation, which generally occurs in anoxic sediments, is not considered very important in freshwater environments (Berman and Bartha 1986; Miskimmin *et al.* 1992).

Many factors can affect the rate of mercury methylation and demethylation including pH, dissolved organic carbon (DOC) concentration, microbial respiration,

water temperature, and lake surface area (Miskimmin et al 1992). These are described further in the following paragraphs.

1.5.1. Effects of microbial respiration

It is now generally accepted that biotic methylation occurs through the action of sulphur reducing bacteria (SRBs) (Gilmour *et al.* 1992; Benoit *et al.* 2003) (Figure 1.2). Sulfate reducers in cultures are effective in methylating mercury, and methylation rates are correlated with the abundance and activity of sulphate reducers (Morel *et al.* 1998). Methylation requires a suitable methyl donor, and methylcobalamin (vitamin B_{12}) is believed to be the only natural methylating agent capable of transferring methyl groups. Methylcobalamin is prevalent in aquatic ecosystems and living organisms, making it the most likely methyl source for environmental mercury methylation (Ridley *et al.* 1977; Ullrich *et al.* 2001). In some species of SRBs (*Desulfovibrio desulficans*) the process is known to be enzymatically mediated, while in others the pathway is uncertain.

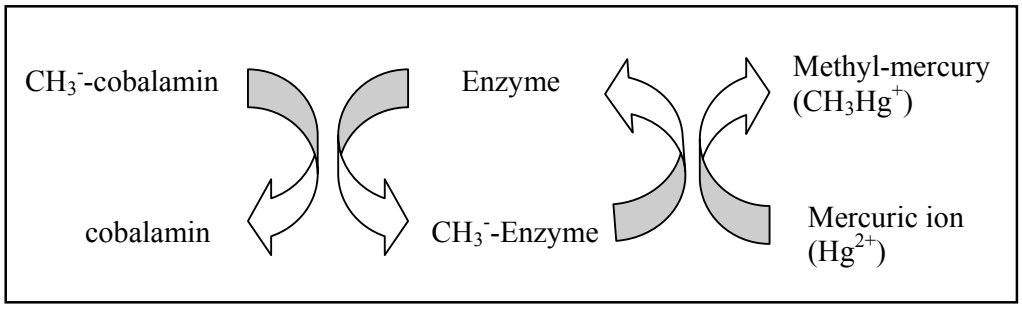


Figure 1.2. Diagrammatic representation of mercury methylation in *Desulfovibrio desulficans* as described in Choi *et al.* (1994).

Methylmercury can be lost through microbial demethylation and photo-degradation. Methylmercury degradation occurs predominantly through bacterial demethylation in sediments and the water column of freshwater lakes. The accepted mechanism of microbial demethylation is cleavage of the carbon – mercury bond by the organomercurial lyase enzyme, resulting in methane and Hg^{2+}. The Hg^{2+} is then reduced to Hg^0 by the mercuric reductase enzyme (Ullrich *et al.* 2001).

Photolytic degradation is an abiotic decomposition process; the reaction is first order with respect to MeHg concentration and the intensity of solar radiation (Sellers *et al.* 1996). Sellers *et al.* (1996) found that MeHg is photolytically

decomposed in surface waters, and that it is potentially the dominant removal pathway for MeHg in epilimnetic freshwater systems. This process on its own may or may not produce Hg^0. Microbial demethylation dominates over photolytic decomposition in deeper water where less light is able to penetrate.

1.5.2. Effects of pH

Mercury concentration in fish is highly correlated with water pH (Cope *et al.* 1990; Grieb *et al.* 1990; Miskimmin *et al.* 1992). Acidic conditions favour mercury methylation in the water column and at the sediment-water interface (Ullrich *et al.* 2001). The pH of the water is considered most influential at the sediment-water interface where lower pH leads to reduced binding of inorganic mercury to DOC, allowing for methylation to occur more readily. Microbial activity, which is correlated with mercury methylation, is greatest near the sediment-water interface, and this activity may be enhanced in low pH waters (Winfrey and Rudd 1990; Miskimmin *et al.* 1992; Schuehammer and Graham 1999). Acidic conditions may favour the activity of microbial species or the operation of biochemical pathways effective at mercury methylation (Lange *et al.* 1993).

As pH decreases, the concentration of MeHg increases in both drainage and seepage lakes (Miskimmin *et al.* 1992). A decrease in pH (a reduction from 7.0 to 5.0 had the greatest effect) alters the protonation of anionic moieties resulting in desorption of metals from DOC and other particulates in the water, rendering the metals available for methylation (Miskimmin *et al.* 1992).

Low pH also favours the production of monomethylmercury over dimethylmercury (McMurtry *et al.* 1989). Dimethylmercury is volatile and is readily lost to the atmosphere from the water surface. Therefore, as lake water becomes acidified and transformation shifts to monomethylmercury production, the waters retain more mercury. Monomethylmercury is stable in water and readily bioaccumulated in aquatic organism tissues, potentially leading to a greater risk of toxicity due to resulting higher tissue concentration (Cope *et al.* 1990; Lange *et al.* 1993; Suns and Hitchin 1990).

Finally, in acidic conditions MeHg can be remobilized from sediments back into the water column as it dissociates from particulate matter in the sediment (Suns and Hitchin 1990). Conversely, at higher pH sediments are more likely to act as a sink than a source for MeHg (Cope *et al.* 1990).

1.5.3. Effects of dissolved organic carbon

Dissolved organic carbon may originate autochthonously, as extracellular products of plants, animals and microbial metabolism, or allochthonously from terrestrial soils and plants (Choi *et al.* 1998). Interactions between DOC and mercury are complex. Dissolved organic carbon can enhance or retard methylation of mercury, serve as a carrier to transport mercury into a water body, or compete with other binding sites for inorganic and organic mercury species. Dissolved organic carbon complexes with heavy metal ions in natural waters (Cope *et al.* 1990; Snodgrass *et al.* 2000). This binding of ionic Hg^{2+} and MeHg decreases the rate of mercury methylation (Winfrey and Rudd 1990). Binding is more pronounced at neutral pH (Ullrich *et al.* 2001). As pH rises, the fulvic acid component of DOC becomes more available for complexation, and the metal ions become less bioavailable because both the OH^- and fulvic acids are competing to form complexes with the metal ions. Watras *et al.* (1995) reported that methylation of Hg^{2+} decreased when DOC was added to the water column. They hypothesized that the observed decrease in methylation was due to a decrease in availability of Hg^{2+} because of its increased binding with DOC. Grieb *et al.* (1990) found a positive correlation between mercury in fish tissue and DOC in drainage lakes, but a negative relationship in seepage lakes.

Some forms of DOC are known sources of decomposable carbon for microbial populations. Therefore, observed increases in MeHg concentration with increasing levels of DOC are attributed to the stimulating effect of organic nutrients on microbial methylating activity (Ullrich *et al.* 2001). Alternatively, it is possible that at high concentrations DOC may inhibit methylating bacteria (Miskimmin *et al.* 1992). Dissolved organic carbon might also decrease methylation rates through complexation with inorganic mercury, thereby decreasing bioavailability. Lastly, in the presence of sunlight, DOC may facilitate the reduction of Hg^{2+} to Hg^0, resulting in dissolved gaseous mercury that can evade from the lake surface resulting in an overall decrease in mercury in that lake (Sellers *et al.* 1996; Zhang and Lindberg 2001). Thus, the precise role of DOC in mercury methylation remains ambiguous.

Dissolved organic carbon and pH interact in a variety of ways that affect the balance of total and MeHg. High DOC (10.5 mg/L) and low pH (<6.0) favour methylation of inorganic mercury over gaseous evasion in aquatic cycling of mercury (Miskimmin *et al.* 1992; Watras *et al*. 1995). In brown water circumneutral

lakes, where in-lake methylation is inhibited by high DOC concentrations, terrestrial inputs of MeHg may be most important (Miskimmin *et al.* 1992). At low pH, it appears that mercury has a lower affinity for DOC allowing it to be methylated more readily.

1.5.4. Effects of temperature and lake surface area

The ratio between methylation and demethylation rates is strongly temperature dependent (Bodaly *et al.* 1993; Ramalal *et al.* 1993). Therefore, any positive correlation between fish mercury concentration and water temperature may be due to the positive influence of temperature on the relative rates of methylation and demethylation. Ramalal *et al.* (1993) found that the rate of mercury methylation was greatest in the epilimnion during summer stratification and the demethylation rate was greatest in the hypolimnion during winter stratification. It is possible that with increasing temperature the rate of methylation increases faster than the rate of demethylation, increasing net methylation. Similarly, methylation rates are higher in warmer epilimnetic sediments than in colder hypolimnetic sediments (Ramalal *et al.* 1993). This is most likely due to the effect of temperature on microbial respiration rates (Ullrich *et al.* 2001). Temperature in turn is related to lake surface area; small lakes are generally shallower, and typically respond more quickly to atmospheric temperature, making them both warmer in the summer and cooler in the winter. This greater temperature range can lead to higher rates of methylation relative to demethylation (Bodaly *et al.* 1993). Variation in surface water temperature is generally greater in shallow rather than deep waters. Similarly, temperature variation is greater in small rather than large lakes, and in the shallow nearshore rather than deeper offshore.

1.5.5. Effects of watershed size and characteristics

Watershed size and characteristics can also affect methylation. Watersheds are recognized as sites of mercury methylation and major sources of mercury inputs to lakes (St. Louis *et al.* 1993). The amount of mercury entering a lake depends on many features of the watershed including vegetation, presence of wetlands, and seasonal occurrences such as spring run-off and flooding (Iverfeldt and Johansson 1988). While wetlands are associated with high methylation rates, their contribution to the mercury budget of a lake depends on their flow rates; peat lands, with low run off rates due to their high water retention, can have relatively small contributions to

mercury inflows (Iverfeldt and Johansson 1988). However, decreased lake size may also result in a greater effect from allochthonous inputs of organic matter by increasing the proportionate flux of wetland derived materials compared to total lake volume (Greenfield *et al.* 2001).

Wetlands are an important source of MeHg. Wetlands, with their large amounts of organic matter, are areas of microbially mediated methylation of inorganic mercury (Berman and Bartha 1986; Choi *et al.* 1998). Upland catchments with good drainage are sites of MeHg retention or demethylation, while catchments with poorer drainage (i.e. wetland areas) are sites of net MeHg production. This may provide an explanation for the high mercury concentrations in fish taken from lakes that are high in colour because they receive water from poorly drained wetlands (Choi *et al.* 1998). The strongest correlations seen between fish mercury levels and relative wetland area are found in circumneutral drainage lakes (Greenfield *et al.* 2001).

The DOC concentration of wetlands is important in affecting mercury transformations. Complexation of DOC with inorganic mercury decreases bioavailability for methylation. Dissolved organic carbon can also increase microbial respiration by acting as a source of decomposable carbon, increasing the production of MeHg. This effect is thought to be small since DOC stimulates abiotic methylation, which is thought to be less important when compared with biotic methylation. Lastly, increased DOC can also increase demethylation rates (Iverfeldt and Johansson 1988).

1.6. Mercury bioaccumulation and biomagnification

Bioaccumulation of mercury is the process by which organisms accumulate mercury at greater than background concentrations as a result of both food intake and uptake from their surrounding environment, e.g. water and the gills. Mercury bioaccumulation in fish tissues can be affected by other factors including aspects of fish biology (size, age, species, and diet), water chemistry (pH, DOC, etc.), and physical aspects of a lake (surface area, watershed area, and watershed characteristics).

Biomagnification of mercury is the increase in its concentration in each successive trophic level in the food chain. Adult lake trout (*Salvelinus* namaycush) can be piscivorous, feeding predominantly on species including lake whitefish

(*Coregonus clupeaformis*), Arctic grayling (*Thymallus arcticus)*, ninespine stickleback *(Pungitius pungitius*), and slimy sculpin (*Cottus cognatus)*. When such species are unavailable, lake trout can feed one trophic level lower on other organisms including crustaceans, and aquatic and terrestrial insects. In some instances, lake trout can be cannibalistic. (Scott and Crossman 1998). Thus, lake trout can feed over three trophic levels as adults. In contrast, lake whitefish are commonly referred to as benthivores, predominantly consuming insect larvae, molluscs, amphipods, gastropods and chironomids. Some lake whitefish do feed on small fish species, however, these are not a primary food source (Scott and Crossman 1998). While mercury concentrations in lake trout tissue in many of Canada's northern lakes approach the 0.5 µg Hg/g limit set by the CFIA, such concentrations are rarely seen in non-piscivorous species including lake whitefish (Evans *et al.* 2005). Methylmercury is the form of mercury that is preferentially biomagnified and the proportion in predaceous species near the top of the aquatic food chain including lake trout is 95 to 99% of THg (Grieb *et al.* 1990). The rate of biomagnification of mercury is controlled through diet, and can be measured through the use of stable isotope analysis techniques which will be discussed later.

1.6.1. Fish mercury levels and fish biology

Various aspects of fish biology including species, age, size, and diet can affect bioaccumulation rates. Different species of fish accumulate mercury at different rates. Piscivorous species such as lake trout acquire more mercury than non-piscivorous species because they feed higher in the food chain, and are therefore more affected by biomagnification.

Many studies have shown a positive association between tissue mercury concentration and both age and size in various species of piscivorous freshwater fish (Grieb *et al.* 1990; Bodaly *et al.* 1993; Lange *et al.* 1993). Mercury concentrations in fish species within a given body of water generally increase with increasing age and body size (both length and weight) (Lange *et al.* 1993; Weiner and Spry 1996). In the NT, piscivorous fish including lake trout, northern pike (*Esox lucius*), and walleye (*Sander vitreus)* in most lakes do not approach the 0.5 µg Hg/g CFIA consumption guideline until they approach 10 years of age (Evans and Lockhart 2001).

Feeding habits affect mercury levels in aquatic organisms (Weiner and Spry 1996). Many studies have provided evidence of mercury bioaccumulation from lower to higher trophic levels in both lacustrine and marine food webs (Cabana and Rasmussen 1994; Kidd *et al.* 1995; Power *et al.* 2002). Power *et al.* (2002) found that mercury concentrations increase by a factor of 5.4 (weighted average) between trophic levels. Higher contaminant concentrations have been correlated with top predators in longer food chains (Kidd *et al.* 1995; Vander Zanden and Rasmussen 1996). Some studies have found that biomagnification rates are greater in more northerly ecosystems compared to lakes in the south, although the mechanisms are not well understood (Kidd *et al.* 1995; Power *et al.* 2002).

1.6.2. Fish mercury levels and water chemistry

Water chemistry and nutrient variables can affect bioaccumulation. Low pH can increase gill permeability and decrease growth rate, therefore decreasing biomass of fish in low pH (<6.0) lakes. The end result can be elevated MeHg concentrations in fish even though MeHg concentrations in the lakes are not high (Winfrey and Rudd 1990).

Dissolved organic carbon inhibits trans-gill transport of MeHg through complexation in the aqueous medium, thereby decreasing MeHg uptake and concentration in fish organs (Choi *et al.* 1998). The complexation of MeHg or inorganic mercury to DOC increases as pH or the amount of DOC increases in solution. As pH increases, fulvic acid becomes more available for complexation, and the mercuric ion becomes less available because OH^- competes with fulvic acid for the mercuric ions (Choi *et al.* 1998). Therefore, in the presence of high DOC concentration in the water, passive uptake of mercury through the gills is decreased, further increasing the proportion of the body burden of mercury acquired through diet. Low or acidic pH was highly correlated with mercury concentration in fish from Wisconsin, Massachusetts, Florida and Sweden (Lee and Hultberg 1990; Lange *et al.* 1993; Watras *et al.* 1998; Rose *et al.* 1999).

1.6.3. Fish mercury levels and physical variables of their lake environments

Metabolic rates of fish are affected by temperature. Increased temperature in the surrounding water increases metabolic rate, leading to increased uptake of food and water, through both the mouth and gills. This, in turn, leads to an increased rate of MeHg uptake (Bodaly *et al.* 1993). Lake size also may be important because

smaller lakes tend to be shallower and therefore warmer in summer than larger, deeper lakes. For example, Bodaly *et al.* (1993) found that mercury concentrations in piscivorous, planktivorous and omnivorous fish were inversely related to lake surface area in northern Ontario lakes: this relationship was attributed to the effect of lake area on temperature. Fish growth rates are affected by crowding; competition among lake trout for food sources is likely greater in smaller than larger lakes. Crowding could result in decreased growth rates and increased mercury concentration in tissues relative to faster growing trout where growth biodilution of mercury could occur (Stafford and Haines 2001). Crowding could also occur if smaller lakes were less intensely fished than larger lakes.

1.7. Previous research on mercury accumulation in northern Canada

While there have been many studies investigating mercury levels in fish, few have been conducted in northern Canada and most were limited in their extent (Shilts and Coker 1995; Stephens 1995). These studies identified high mercury levels in piscivorous fish in a number of small, remote lakes and resulted in the first consumption advisories for freshwater fish in the NT. These findings were confirmed by Lockhart (1998) in a broader survey of lakes in the NT where fish were commonly found to have mercury levels above 0.5 µg/g. This led to expanded studies, supported by the Northern Contaminants Program (NCP) of the Indian and Northern Affairs Canada, and conducted by Environment Canada and Fisheries and Oceans Canada (DFO) research scientists investigating mercury levels in fish and the primary factors affecting these levels. The NCP does not support research in the northern provinces, however, the Toxic Substance Research Initiative (TSRI), which ran from 2000-2004, included a study investigating mercury (and persistent organic pollutants) in lake trout from the northern provinces (Muir *et al.* 2001). This thesis is based on data collected during these two studies with additional data generated from two smaller studies, i.e., the Northern Ecosystem Initiative (NEI) funded study and a Sahtu Renewable Resources Board (SRRB) funded project. Highlights of these programs and their relevance to this thesis and its objectives are presented below.

1.7.1. The Northern Contaminants Program

In the mid 1990s, the first evidence of high (> 0.5 µg Hg/g) mercury levels in lake trout, northern pike, and walleye living in small remote lakes in the NT was discovered by Stephens (1995). Subsequent studies conducted as part of routine stock assessment investigations confirmed that elevated mercury levels in predatory fish were widespread in the NT (Lockhart 1998). In many areas, the fish tissue mercury concentration exceeded both the 0.2 µg Hg/g FCFG, and the 0.5 µg Hg/g CSFG (Evans and Lockhart 2001). This lead to human health concerns because fish comprise an important portion of the human diet in this region. Also, from a socio-economical perspective, high mercury levels are problematic to the 'First Nation' communities in the area. These communities depend on commercial fisheries for income, for maintaining cultural values, and for tourism, which is heavily reliant on the sport-fishing industry (Evans and Carpenter 1997). However, while mercury concentrations are high in piscivorous fish in some lakes, there are lakes where levels are quite low. While there were many possible explanations for these relatively high mercury concentrations in fish, without additional study, it was impossible to determine which were the key factors affecting these high mercury concentrations in fish in northern lakes.

The NCP-funded study undertaken by Dr. M. Evans from Environment Canada and Dr. L. Lockhart from the DFO, was designed to investigate factors involved with spatial variations in mercury concentration in fish tissue in the Mackenzie River Basin (MRB). The species of fish inhabiting the upper trophic levels included in the studies were northern pike, walleye, and lake trout. Lockhart studies focussed on measuring mercury concentrations in fish as part of additional stock assessment studies and further characterizing the extent of the lakes of concern. Recently this work was synthesized and broadened to include lake trout (and other species) from all Canadian Territories. Lockhart *et al.* (2005) determined that lake trout in sampling sites in the NT had mean concentrations of mercury that were above the 0.2 µg Hg/g FCFG concentration, but just below the 0.5 µg Hg/g CSFG level. However, a quarter of the sites had mean tissue concentrations above 0.5 µg Hg/g. As expected, these studies showed large spatial variation in mercury concentrations in predatory fish in the NT. On a more positive note, the study found that the presence of mercury in excess of 0.5 ug/g in freshwater fish is largely

restricted to piscivorous species including lake trout, northern pike and walleye (Lockhart *et al.* 2005). The Evans study began with NCP support in 1998. It incorporated a large number of fish of each species collected from various lakes, including a number of lakes previously sampled as part of the DFO stock assessment studies. An overview of this work is presented in Evans *et al.* (2005).

There were no immediate explanations for the wide variation in mercury concentrations found in fish tissues from the different lakes. The environments in which these lakes are situated are relatively untouched by direct anthropogenic pollution, although they are affected by long-range atmospheric deposition. There are no activities such as mining, pulp and paper mill operations, reservoir creation, or clear cutting, all of which can have impacts on mercury concentration in lakes (Jackson 1991; Tremblay and Lucotte 1997; Garcia and Carignan 2000). Low water pH increases MeHg concentrations in fish tissue (Miskimmin *et al.* 1992), yet most lakes in the area of study are located on glacial tills and are not acidic (Evans and Lockhart 2001). The most important variables leading to higher mercury levels in fish in the NT appear to be fish age and size (Evans and Lockhart 2001). Fish in the north are characterized by slow growth and old age. Cold climate leads to lower fish growth rates than in warmer and more southern latitudes. In addition, light fishing pressures due to low human population densities combined with few natural predators, allows the fish to survive to great age (greater than 23 years, Scott and Crossman 1998). Mercury concentrations in piscivorous fish do not approach 0.5 µg Hg/g until these fish reach ca. 10 years of age (Evans and Lockhart 2001). Lake size also appeared important with mercury levels tending to be high in fish in smaller lakes (Evans *et al.* 2005).

Despite these general understandings, it was highly desirable to obtain more quantitative information on the factors affecting mercury in NT lakes with the goal of developing predictive models to identify lakes with predatory fish populations having high tissue mercury concentrations. The NCP project has recognized the importance of studying the factors affecting lake-to-lake variation of mercury levels in predatory fish (Evans and Lockhart 2001). However, no effort has been made to use the measured variables to determine a model to predict mercury concentration in fish tissues. Moreover, the relationship between mercury concentrations in fish, the physico-chemical features of their aquatic environment and mercury levels in the food web have yet to be thoroughly investigated.

The geographic regions covered by the study lakes in the Mackenzie River Valley, NT, are highly variable, from high elevation mountain lakes to lowland lakes. The study area also is much larger than those previously studied, covering over 500,000 km^2, with a wide range in both latitude and climate. The lakes and their watersheds also vary in size from very small (Mirror Lake) to very large (Great Bear Lake). Thus, this data set provided an excellent opportunity to investigate the factors or combination of factors responsible for variations in mercury levels in the MRB. Of particular interest is the development of predictive models to estimate tissue mercury concentrations in lake trout in the many unstudied lakes in the region.

1.7.2. Toxic Substance Research Initiative (#236) project

The Toxic Substance Research Initiative (TSRI) project was titled "Biomagnification of Persistent Organic Pollutants and Mercury in Canadian Freshwater Subsistence Fisheries and Food Webs". The study was designed to determine levels of persistent bioaccumulative toxic substances, including mercury, in top predator fishes and their food webs from lakes across northern Canada ranging from Alberta to Labrador. Because the study area covered a vast region, it provided spatial information on mercury levels and their bioaccumulation in aquatic biota with a focus on lake trout. The study was initiated because many of the lakes supported important 'First Nations' subsistence fisheries and, relative to the Great Lakes and the Arctic, the lakes in this geographic region were understudied with respect to persistent organic pollutants (POPs) and mercury.

The TSRI-funded study resulted in the first large multi-lake dataset which included levels of persistent organic pollutants (POPs) and mercury in food webs of lake trout lakes, combined with information on water chemistry and food webs (Muir *et al.* 2001). This information could be compared with findings in lake trout lakes in the NT to investigate the factors responsible for spatial variations in mercury levels in lake trout. This thesis bases such comparisons only on the northern Alberta and Saskatchewan (NAS) TSRI data, i.e; lakes part of the Mackenzie River and Churchill River basins.

Initially it was expected that mercury levels would be higher in lake trout in NAS than NT lakes because of their closer proximity to anthropogenic sources of mercury and their warmer lake temperatures (summer). However, when the data were compared, the mercury concentrations in fish species in NAS lakes were lower

than the concentrations found in fish from the NT lakes (Evans *et al.* 2005). While some of these differences were related to fish age, i.e., lake trout were considerably younger in the NAS than they were in the NT lakes, factors such as bioaccumulation through the food web and fish growth rates were not considered. Differences in food chain length or biomagnification rates may differ as a function of latitude; growth rates may also differ with concomitant consequences to mercury levels in lake trout. Thus, the combined TSRI and NCP data sets provided an excellent opportunity to explore these questions.

1.8. Thesis objectives

The main goal of this thesis was to determine the factors that influence mercury accumulation in piscivorous fish in a northern freshwater environment using lake trout as the study species. In addition there were a number of smaller objectives designed to evaluate the effects of specific factors on mercury accumulation.

The objectives of this study were:

1. To quantify the variables of most importance in affecting mercury levels in lake trout in lakes in the NT, and to develop a predictive model for mercury bioaccumulation in lake trout in the NT, Canada, that could be used to estimate mercury concentration in lake trout muscle tissue;

2. To determine the basic features of lake trout feeding using stable isotope studies, and use these data to investigate regional differences in mercury biomagnification rates in lakes in the NT and NAS; and

3. To determine the effects of fish age and growth rate on the concentration of mercury in muscle tissue of lake trout and to investigate differences between lake trout populations in NAS.

2.0 AN INVESTIGATION OF THE PHYSICAL, CHEMICAL, AND BIOLOGICAL FACTORS AFFECTING MERCURY CONTAMINATION IN LAKE TROUT, *SALVELINUS NAMAYCUSH*, IN THE MACKENZIE RIVER BASIN

2.1. Abstract

Mercury is a contaminant of global concern; many industrialized areas of Canada and the United States have issued consumption advisories for different species of fish from mercury-contaminated waters. Mercury concentrations are also high in the tissues of fish from many northern Canadian lakes, far from point sources of anthropogenic pollution; consumption advisories have been issued. While elevated mercury concentrations in aquatic ecosystems are associated with many different variables, the specifics of these associations are unknown in northern Canada. Therefore, a series of lakes in the Mackenzie River Basin was investigated to quantify the combination of factors responsible for variations in mercury concentrations in lake trout (*Salvelinus namaycush*) from northern Canadian lakes. This predictive capability could provide the 'First Nations' people with advice on probable mercury concentrations in the many unstudied lakes. A variety of physical, chemical, and biological variables were measured based on factors previously identified as those related to mercury bioaccumulation in other regions. The resulting model was composed of latitude, fish age, fork length, lake surface area, and the watershed area/lake area ratio, and explained 75.9% ($p < 0.001$) of the variance in tissue mercury concentrations in lake trout.

2.2. Introduction

Mercury bioaccumulates in freshwater and marine food webs. This can lead to high mercury concentrations in fish tissue, particularly in piscivorous fish feeding at upper levels of the food chain, even in lakes relatively undisturbed by direct anthropogenic inputs (Weiner and Spry 1996). Mercury concentration can increase over six orders of magnitude from water to fish tissue (Watras *et al.* 1995), and therefore, the mercury concentrations in the muscle tissue of large, piscivorous fish

can reach levels that are potentially toxic to human consumers. High mercury concentrations in fish tissues are particularly alarming for people whose diets are high in fish content, including the people of Canada's northern communities. Studies have found high mercury concentrations in piscivorous fish species in many remote lakes in northern Canada that are far from point sources of mercury pollution, from the eastern Arctic, through the Northwest Territories (NT), and into the Yukon Territory (Lockhart 1998; Braune *et al.* 1999, Evans *et al.* 2005).

The bioaccumulation of mercury in aquatic food chains is controlled by many interrelated factors including biological aspects of fish, as well as chemical and physical variables of the aquatic environment the fish inhabit. Variations in these factors lead to large differences in mercury concentrations observed in the tissue of fish from lakes in close proximity to each other. Within a region, mean mercury concentrations in piscivorous fish species such as lake trout can vary five times or more between lakes, even following standardization for fish age and size (Bodaly *et al.* 1993). Physical-chemical factors which facilitate the transformation of inorganic mercury to methylmercury (MeHg) are important, since 95 to 99% of total mercury (THg) in fish tissue is MeHg (Grieb *et al.* 1990). Chemical variables previously associated with increases in MeHg concentration in drainage lakes include high dissolved organic carbon (DOC) (Watras *et al.* 1998), low pH (Grieb *et al.* 1990; Miskimmin *et al.* 1992; Watras *et al.* 1998; Scheuhammer and Graham 1999); temperature is the primary physical variable with methylation rates increasing with increasing temperature (Ramlal *et al.* 1993).

Other physical variables that can affect mercury concentration in lake water are lake surface area and watershed, or drainage area. Lake surface area can affect mercury concentrations in fish tissue in a number of ways. Smaller lakes increase in temperature more quickly in the summer months, allowing for increased MeHg formation (Bodaly *et al.* 1993; Ramalal *et al.* 1993). Increasing water temperature also affects fish metabolic rates, increasing both feeding and gill ventilation (Bodaly *et al.* 1993), which can lead to greater mercury uptake by the fish. Watershed area can play a large role in mercury biomagnification (McMurtry *et al.* 1989) because watersheds are a source of mercury inputs to a lake, together with atmospheric inputs (Iverfeldt and Johansson 1988). Watersheds can also provide favorable conditions for increased inorganic mercury to MeHg transformation if they contain shallow

water sites including marshes and wetlands. These ecosystems are rich in DOC and are often warmer than the lakes into which they drain.

Biological variables including fish length, weight, age, and diet (Grieb *et al.* 1990; Power *et al.* 2002; Evans *et al.* 2005) affect their mercury concentrations over time because mercury is readily taken up by fish through both from their diet and surrounding water, but is poorly eliminated (Ulrich *et al.* 2001). This allows mercury to accumulate in fish tissues over time reaching high concentrations in old and large fish. Fish diet can be investigated through the use of stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes (Kling *et al.* 1992; Vander Zanden and Rasmussen 1996; Power *et al.* 2002). Nitrogen isotopes can provide information on trophic level, including whether the fish diet is of piscivorous or omnivorous nature and on the length of the food chain in a given lake (Cabana and Rasmussen 1994). Carbon isotopes can be used to determine whether a fish diet is composed of foods of benthic or pelagic origin (Hecky and Hesslein 1995). Recent studies suggest that allochthonous, benthic and littoral zone sources of carbon decrease with increasing lake surface area (Perga and Gerdeaux 2004).

The manner in which these factors interact to influence mercury concentrations in northern Canadian lakes is not yet well understood. This prevents researchers from making knowledgeable inferences of mercury concentrations in previously unstudied lakes. There are hundreds, if not thousands, of unstudied lakes in the NT from which 'First Nations' people harvest fish for consumption. It is not practical to investigate mercury concentrations in fish from all these lakes.

The objective of this study was to investigate the primary factors, or combination of factors, responsible for high mercury concentrations in lake trout from 17 lakes located in the Mackenzie River Basin (MRB) in the NT. Several biological, physical, and chemical variables were investigated to determine the primary variables influencing mercury concentrations in lake trout. Biological variables included fish fork length, weight, age, $\delta^{13}C$, $\delta^{15}N$ and mercury concentration in zooplankton. Physical variables were lake latitude, lake depth, lake surface area, watershed area to lake surface area ratio, and water temperature. Chemical variables evaluated were pH, and DOC, total phosphorus (TP), chlorophyll *a* (Chl*a*), dissolved oxygen (DO), THg, MeHg and specific conductivity in water. Relationships between these biological, physical, and chemical variables were then explored further through multiple regression analyses to determine the primary

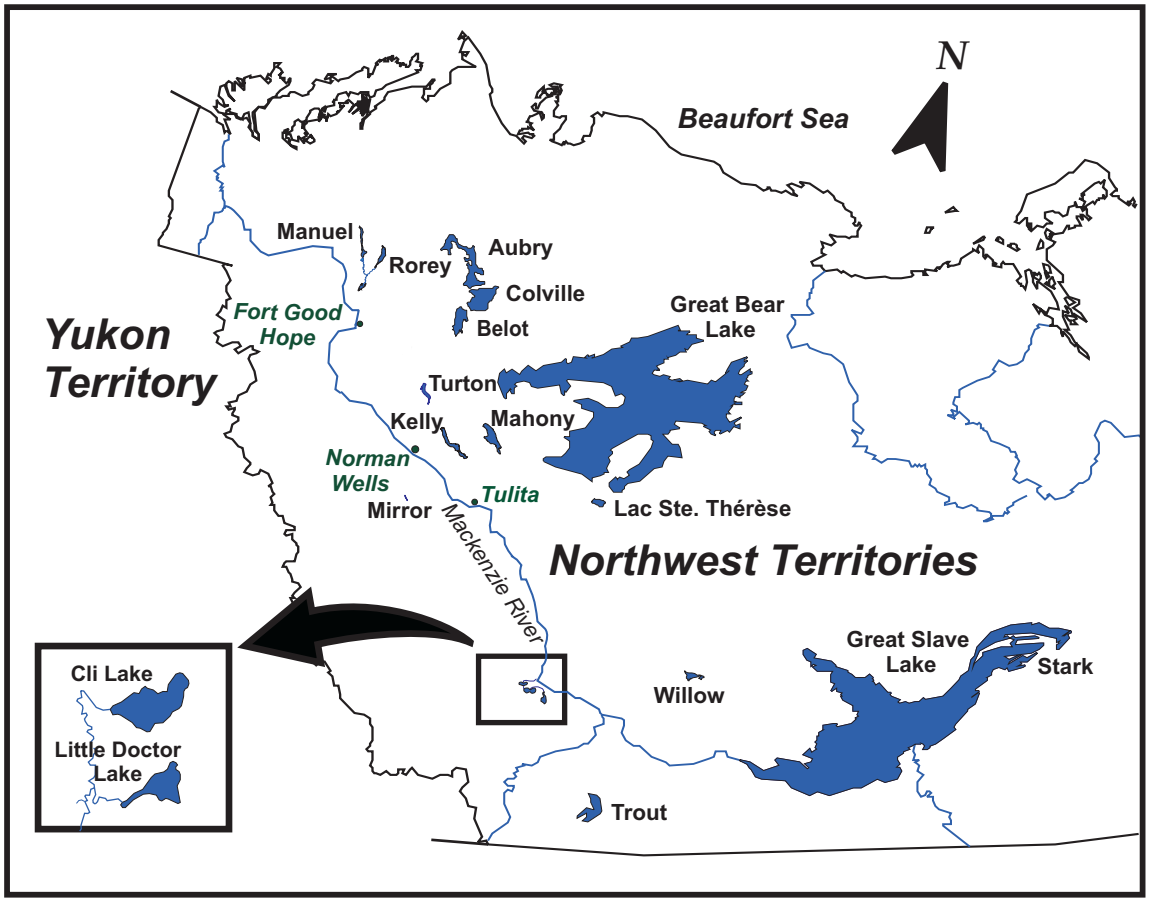


Figure 2.1. Map of the Northwest Territories, Canada, showing the 17 study lakes

Table 2.1. Geographic location, surface area, and watershed area of the 17 study lakes in the Mackenzie River Basin, Northwest Territories, Canada.

Lake	Latitude (deg)	Longitude (deg)	Lake surface area (km^2)	Watershed area (km^2)	Lake surface area/ Watershed area
Aubry	67° 24’ N	126° 27’ W	390.70	781.40	2.00
Colville	67º 10’ N	126º 00’ W	447.94	904.84	2.02
Manuel	66° 58’ N	128° 54’ W	51.81	489.60	9.45
Rorey	66° 55’ N	128° 24’ W	59.89	432.41	7.22
Belot	65° 53’ N	126° 16’ W	304.31	648.18	2.13
Great Bear	65º 11’ N	125º 25’ W	31,328.00	115,287.04	5.18
Turton	65° 48’ N	128° 24’ W	47.67	743.65	15.60
Mahony	65° 30’ N	125° 20’ W	181.03	1055.40	5.83
Kelly	65º 23’ N	126º 15’ W	120.82	808.29	6.69
Mirror	64º 51’ N	120º 45’ W	2.96	11.13	3.76
Ste. Thérèse	64º 38’ N	121º 35’ W	113.47	2888.95	25.46
Stark	62º 28’ N	110º 20’ W	273.58	749.61	2.74
Willow	62º 10’ N	119º 08’ W	159.69	1085.89	6.80
Great Slave	61° 30’ N	113° 50’ W	28568.00	985310.32	34.49
Cli	61º 59’ N	123º 18’ W	44.15	123.18	2.79
Little Doctor	61º 53’ N	123º 16’ W	20.54	395.40	19.25
Trout	60º 35’ N	121º 19’ W	507.50	2106.13	4.15

factors affecting tissue mercury concentrations in lake trout. Finally, the data were used to develop a predictive model of mercury concentrations in fish which could then be applied to unstudied lakes. This model was further simplified, requiring fewer measured variables, and with potential to be used as a screening tool by First Nation communities and others to determine mean mercury levels in lake trout in unstudied lakes. Of particular interest was the capability to predict lakes which contained lake trout with mercury levels in excess of 0.5 µg Hg/g.

2.3. Materials and Methods

2.3.1. Study area

The 17 lakes included in this study are all located in the NT, Canada, in the MRB. The lakes lie along both sides of the Mackenzie River from Trout Lake in the south to Aubry Lake in the north. The topography and geology of this region varies widely, including the Mackenzie and Franklin mountains, as well as the Horn Plateau and numerous other isolated hilly areas (http://www.ccea.org/ecozones/index.html, 09/13/2006). Stark and Great Slave lakes lie partly on the Canadian Shield, while all remaining lakes lie on paleozoic deposits. The climate is sub arctic with mean monthly temperatures ranging from approximately -25°C in January to 17°C in July, with a mean annual temperature of approximately -4°C (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/stnselect_e.html, 01/18/2005). The study lakes vary in total surface area, from Mirror Lake, which is 3 km^2, to Great Bear Lake, which is 31,328 km^2 (Figure 2.1 and Table 2.1). All lakes are located below the tree line, with northern boreal tree species including white spruce, (*Picea glauca*), birch, (*Betula papyrifera var. papyrifera*), aspen, (*Populus tremuloides*), and jack pine, (*Pinus banksiana*) being found. In low-lying areas, the trees give way to muskeg. All lakes are drainage lakes (i.e. those fed primarily by streams and with outlets into streams or rivers), and are normally covered with ice from October until mid-June.

2.3.2. Sampling methods

2.3.2.1. Fish collections

Fish sampling from the NT lakes, except for Ste. Thérèse, Great Slave, Great Bear, Stark and Trout lakes, was conducted from 1996 to 2002. These lakes were sampled as part of the stock assessment studies conducted by the Department of Fisheries and Oceans (DFO), Hay River (Lockhart *et al.* 1998; Evans and Lockhart 2000, 2001; Lockhart *et al.* 2001; Stewart *et al.* a, b, 2003). Ste. Thérèse, Great Slave, and Great Bear lakes were sampled over the same time period by M. Evans from National Water Research Institute (NWRI), Saskatoon (Evans *et al.* 2005). Although the fish collections were conducted over a number of years, Lockhart *et al.* (2005) determined that there was no systematic change in mercury concentrations or growth rates in fish over the study years.

Fish collected during stock assessment studies were generally caught in the late winter when lakes were accessible by snowmobile, and freezing temperatures provided natural means of refrigeration for the large number of fish collected. Fish were captured using multi-panel gillnets set on the lake bottom. These panels were of three mesh sizes (89 mm, 114 mm, 140 mm stretched measure) and measured either 45.5 m or 91.4 m in length and either 1.83 or 3.66 m in depth. The short, shallow nets were generally set in small, shallow lakes and *vice versa*. Most nets were set overnight and pulled the following day (Stewart *et al.* a, b, 2003). Fish from Great Slave, Great Bear, Ste. Therese, Stark and Trout lakes were caught by local community members as part of their subsistence fisheries. Nets generally were set in late autumn to meet demands for submission of samples to the analytical laboratory in the same fiscal year (April 1 to March 31). Gill nets were composed of 140 mm mesh netting.

Fish round weight, fork length, age and sex were determined for the stock assessment studies and for Stark and Trout Lake fish. For Great Slave, Great Bear, and Ste. Therese lakes fish, fish were frozen whole and shipped to NWRI, Saskatoon for processing. Otoliths were removed for later age analyses and sections of dorsal muscle were removed, placed in a sterile Whirl-Pak® bag, and frozen for mercury and stable isotope analyses. Muscle tissue and aging structures were sent to DFO, Winnipeg, for mercury analyses. Stable isotope analyses were performed at NWRI, Saskatoon.

2.3.2.2. Zooplankton sampling

Zooplankton samples were collected at 11 of the 17 lakes during the summers of 2001-2003. Access to many of the lakes was done by Twin Otter Floats, but the high costs associated with float plane charters did not allow all lakes to be sampled during the period in which funding was available. Zooplankton were collected during summer limnological sampling using a # 10 mesh plankton net (156 μm) outfitted with a # 10 mesh plankton bucket (cod end). In areas where there was low plankton biomass (ex. Great Bear Lake) a #20 mesh (76 μm) net and cod end were used. Zooplankton nets were towed in open water areas in close proximity to the deepest part of the lake, (at the same site as other limnological sampling was conducted). For larger lakes (Great Bear, Great Slave, Colville, Stark lakes) sampling was conducted a few kilometres from shore in offshore waters. The net was towed behind a slow moving boat so that the top of the mouth of the net was approximately 1 m below the water surface. Each tow lasted approximately 15 minutes and triplicate samples were collected. Collected zooplankton was deposited into a Whirl-Pak® bag, kept in a cooler with ice packs, and frozen as soon as possible. On return to NWRI, Saskatoon, zooplankton samples were freeze-dried, and sub-sampled for stable isotope and mercury analyses.

2.3.2.3. Mercury in biological tissue

Lake trout and zooplankton samples were individually subsampled (20 mg wet weight (ww) for the fish and >100 mg dry weight (dw) for the zooplankton samples) upon return to NWRI, Saskatoon. These sub-samples were then sent to the DFO in Winnipeg, for THg analyses.

All acids used in the analysis were trace metal analysis grade (concentrated) unless otherwise specified. All water was distilled and deionized. Commercial Atomic Absorption (AA) standards, reagent blanks, and standard reference materials were digested concurrently with the samples. Test tubes used for digestion were 25 x 200 mm Pyrex glass and were washed with 10% nitric acid followed by a deionized water rinse prior to use. The aluminium block heater used for digestions was both time and temperature programmable. Standard reference materials (National Research Council (NRC)) were analyzed following each sample run. Total mercury was analyzed using the 'hot block digestion - cold vapour atomic absorption' method (Hendzel and Jamieson 1976). A small subsample of wet tissue

(0.2 g) was digested with 5 mL of 4:1 sulphuric:nitric acid at 180°C for 12 hours, cooled, and diluted to 25 mL with deionized water. Elemental mercury was released from this solution with a stannous chloride reductant and carried by a stream of air to a model 3200 Mercury Monitor from LDC Analytical for atomic absorption detection (detection limit; 0.01 µg Hg/g ww).

2.3.2.4. Stable isotope analyses

Stable nitrogen and carbon (non-lipid extracted tissues) isotope analyses were conducted by the NWRI stable isotopes laboratory (Saskatoon, SK). Biological tissues were freeze-dried at -60ºC and ground to fine powder with a mortar and pestle. Between 1 and 2 mg of sample was required for analysis. Ground samples were analyzed using a VG-Isochrom Continuous Flow Isotope Ratio Mass Spectrometer (CF-IRMS) from Micromass (a division of Waters Corporation) using methods described in Teece and Vogul (2004). Stable nitrogen and carbon isotope ratios in the biota are expressed in parts per thousand (‰ or per mil) difference from the standard air ($^{15}N/^{14}N$ = 0.00367) and Pee Dee Belemnite ($^{13}C/^{12}C$ = 0.01123) respectively using the following equation:

$$\delta^{15}N \text{ or } \delta^{13}C \text{ ‰} = [(R_{sample}/R_{standard})-1] \times 1000 \quad (2.1)$$

where $R = {}^{15}N/{}^{14}N$ or $^{13}C/^{12}C$.

A laboratory working standard, Pharmamedium, a cottonseed protein of known, constant, stable $\delta^{15}N$ and $\delta^{13}C$ isotope ratios, was run every 5 to 10 samples for both N and C analyses.

2.3.2.5. Water quality

Water quality profiles could be conducted at only 11 of the 17 study lakes due to the logistical and financial restraints associated with accessing these remote locations. Most of the sampling occurred once at each of the lakes during the summer (July to August) of 2001 to 2003. The lakes sampled were Colville, Great Bear, Kelly, Mirror, Ste. Thérèse, Stark, Willow, Cli, Little Doctor, Trout and Great Slave. Cli, Little Doctor and Willow lakes were sampled multiple times over four summers (1999-2002), but there was little variation in water quality over time, so the most recent data available was used. Water quality data collected as part of this study was supplemented with unpublished water quality data collected in 1999 from Manuel, Rorey, Belot, Turton, and Mahony lakes (Stephens G., Indian and Northern

Affairs Canada, unpublished data). These data were limited to pH, conductivity, TP, temperature, and DO.

For the lakes investigated during our program, detailed limnological sampling was conducted at the deepest portion of the lake (determined by bathymetric mapping) or in the most offshore waters that were practical to sample given the size of the lake. Temperature, pH, conductivity, and DO profiles with depth were collected using a Hydrolab H_20 Multiprobe Sonde (Hach Environmental), which was calibrated daily. Measurements were taken every 2 meters beginning at the surface and ending at approximately 1 meter above the lake bottom. Water samples were collected at the surface and at approximately 1 meter from the lake bottom. In some of the deep lakes, water was collected at a third depth (mid-depth or just below the thermocline). These samples were kept cool, first in coolers containing a few ice packs until return to shore, where they were field filtered, and then at the end of the field sampling day, placed in refrigerators until shipped to NWRI in Saskatoon. Total phosphorus (TP), DOC, and Chl*a,* were determined as in Robarts *et al.* (1989). Where there were samples from multiple depths in the same lake, the median is reported.

Water samples for mercury and MeHg analyses were collected using a 'clean hands, dirty hands' protocol (US EPA 1996). This process required three people for proper sampling. One person drove the boat nose into the wind; both clean and dirty hands people donned a new pair of sterile gloves. Dirty hands selected a double-bagged 250 mL sample bottle and opened the outer bag. Clean hands then opened the inner bag and removed the sample bottle, which was completely filled with dilute hydrochloric (HCl) acid. This dilute acid was discarded into the lake and the sample bottle rinsed 2 to 3 times with lake water. From the front of the boat the sample bottle was held approximately an arm's length (20 to 30 cm) below the water surface and slowly filled as the boat moved forward. The sample bottle was then recapped by clean hands and placed back into the inner bag, which was then sealed. Dirty hands then closed the outer bag and returned the sample bottle to the cooler. This process was then repeated for the MeHg sample bottle. These two samples were then preserved with 10 mL 0.2% HCl acid before being shipped to Flett Research Ltd. in Winnipeg, for analyses.

2.3.2.6. Total mercury in water

The method used by Flett Research to analyze water samples for THg followed that of Bloom and Crecelius (1983), which has been adopted by the US Environmental Protection Agency (US EPA) (US EPA 2002). Water samples, received in 250 mL acid-cleaned glass bottles with Teflon lined caps, were preserved with 0.2% HCl. The samples were spiked in the laboratory with bromine chloride (BrCl), a strong oxidizing agent, and allowed to react for at least 8 hours. The residual BrCl was then neutralized by the addition of hydroxylamine hydrochloride to prevent destruction of gold traps. The digested sample was placed into a sparging vessel (bubbler), and the reducing agent stannous chloride was added to reduce Hg II to Hg vapour, or elemental mercury (Hg^0). The elemental mercury produced was bubbled off using N_2 gas and collected on a gold trap. The gold trap was heated to approximately 400°C, and argon carrier gas was passed through and the elemental mercury was measured by atomic fluorescence spectroscopy. National Institute of Standards and Technology (NIST) related standards were used. The detection limit for 100 ml samples was approximately 0.1 ng Hg^0/L.

2.3.2.7. Methylmercury in water

Water samples, contained in 250 mL acid-washed glass bottles with Teflon lined caps, were received preserved with 0.2% clean HCl to halt any biological activity until analysis could be performed. Samples were first distilled to remove MeHg from any unknown substances present in the water sample. Methylmercury was then ethylated to ethylmercury with sodium tetraethyl borate. Ethylmercury was then purged onto a Tenax® (Scientific Instruments Inc.) trap using N_2 gas. Tenax® traps are made of organic polymer, and have a high affinity for gaseous organics; this affinity is reversible when heat is applied. The traps were dried with nitrogen, and then heating in an argon gas stream (approximately 400°C) that swept the analyte onto a gas chromatograph (GC) column for separation of the ethyl MeHg from other ethylated mercury compounds (Liang *et al.* 1994). The analytes were then passed through a pyrolizer heated to 700 to 800°C where the organic mercury was converted to Hg^0 before entering a cold vapour atomic fluorescence analyser for detection (Horvat *et al.* 1993). Matrix spikes and matrix spike duplicates as well as two process blanks were included with every eight samples. The method detection limit was approximately 0.04 ng Hg^0/L for a 40 mL sample.

2.3.2.8. Lake surface area and watershed area determination

Lake surface area and watershed area of the 17 study lakes were determined from National Topographic Data Base (NTDB) digital maps, 1:50,000 and 1:250,000 scale. Lake area was calculated by isolating individual lake polygons. The areas were derived from these polygons and the ArcGIS visual basic scripting tool. Files were accessed and manipulated using the ArcGIS suite, v8.x, an Environmental Systems Research Institute (ESRI) product (2000).

Watershed area was delineated using the Canadian National Topographic Data Base (NTDB) 1:250K map set. The required digital map sheets were loaded into the ArcINFO 9.x suite of GIS software and a Universal Transverse Mercator; Zone 12 projection was used. All data was geodetically referenced to North American Datum 1983, using the of World Geodetic Survey 1984 ellipsoid.

2.3.3. Statistics

Statistical analyses were performed to investigate the factors affecting mercury levels in lake trout within and between lakes. Once these factors were determined, a series of analyses were performed in order to develop predictive models which could be used to estimate mercury concentrations in an individual fish based on these considerations.

First, the factors affecting mercury concentrations in lake trout in each of the 17 lakes were investigated using linear regression analyses. Variables considered were fish age, fork length, weight, $\delta^{13}C$ and $\delta^{15}N$. Next, linear regression analyses were performed to investigate the factors affecting variation in mercury levels in fish between the 17 lakes. In addition to the five variables already noted, geographic (latitude, lake surface area, watershed/lake surface area), physical (temperature), and chemical (DOC, pH, TP, Chl*a*, THg in water, MeHg in water, and THg in zooplankton) variables were considered. Data normality and variance homogeneity were assessed prior to statistical analyses. Mercury concentration in lake trout and lake surface area was log transformed to stabilize variance. Statistical significance of each of the relationships was determined using *F*-statistics calculated for the regression, and *p*-values. Maximal Type I error for all analyses was set at $\alpha = 0.05$. Data are reported as correlation coefficients.

Next, best subsets regression analysis was used to determine which of these biological, geographic, physical, and chemical variables together described the most

accurate and parsimonious model to estimate mercury concentration in lake trout in a given lake. Model fitness was evaluated through r^2 and C_p statistics. Best subsets analysis was used because it allows the user to decide between a variety of possible models based on the r^2 and C_p statistics. If the model is an appropriate one, the C_p value should approach p+1, where p is the number of predictors. While the r^2 value is a measure of model fit, the C_p statistic is a measure of model fit and frugality. Unlike stepwise regression, best subsets regression places variable selection back in the hands of the researcher, who understands underlying theory, variables, and resources involved (King 2003). Finally, multiple regression analyses were calculated using the variables selected through best subsets regression. Variables associated with individual lake trout with large standardized residuals were classified as statistical outliers and were removed from the equation to strengthen the relationship.

Once the predictive models were developed, their predictive capability was assessed by using the model to estimate mercury concentrations in lake trout from Rae (64° 10'N, 117° 20'W) and Faber (63° 56'N, 117° 15'W) lakes. These two small lakes are located on the Canadian Shield north of Yellowknife, and mercury concentrations were determined in 2000 (Stoddart *et al.* 2001). Differences between measured and predicted mercury concentrations in Rae and Faber lakes were explored using one-way analysis of variance (ANOVA). All statistics were performed using Minitab 13 statistical software (Minitab, Inc. 1999).

2.4. Results

2.4.1. Lake trout statistics

All detailed statistics on lake trout collected for this study can be found in Table 2.2, and raw data for all fish sampled are found in Appendix A. The mean age of lake trout among all the lakes ranged from 9.3 years in Aubry Lake to 24.9 years in Lac Ste. Thérèse, with a mean and standard deviation of 15.2 ± 4.4 years. Mean fork length of the lake trout from all lakes ranged from 468.9 mm in Mirror Lake to 736.9 mm in Great Bear Lake, with a mean and standard deviation of 593.6 ± 72.5 mm. Mean weight ranged from 1455.6 g in Rorey Lake to 4166.5 g in Mahony Lake, with a pooled mean and standard deviation of 2528.2 ± 1132.6 g from all lakes.

Table 2.2. Mean values ± standard deviation of biological variables including lake trout age, fork length, weight, carbon[13] isotopes, nitrogen[15] isotopes and total mercury (THg) concentration in samples of dorsal muscle tissue, in lake trout, *Salvelinus namaycush*, caught from 17 study lakes in the Northwest Territories, measured on a per lake basis. Raw data for all lake trout sampled are found in Appendix A.

Lake	n	Age (years)	Fork length (mm)	Weight (g)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	THg (µg/g ww)
Aubry	20	9.3 ± 2.1	592.0 ± 70.0	2422.0 ± 851.2	-24.14 ± 2.17	11.07 ± 0.79	0.27 ± 0.09
Colville	39	11.3 ± 3.1	576.4 ± 36.8	1955.0 ± 455.4	-25.65 ± 1.33	16.18 ± 0.44	0.21 ± 0.07
Manuel	19	14.5 ± 4.5	484.4 ± 30.0	1380.0 ± 240.3	-28.45 ± 1.20	11.51 ± 0.48	0.30 ± 0.06
Rorey	20	14.4 ± 3.4	525.3 ± 36.0	1455.5 ± 251.2	-28.28 ± 1.10	11.31 ± 0.53	0.44 ± 0.17
Belot	20	13.3 ± 4.9	609.5 ± 54.6	2866.5 ± 669.0	-23.75 ± 1.83	10.79 ± 0.60	0.20 ± 0.08
Great Bear	10	19.9 ± 5.1	736.9 ± 122.5	4086.4 ± 1587.6	-27.58 ± 1.49	13.29 ± 0.97	0.35 ± 0.31
Turton	20	11.4 ± 3.3	558.5 ± 36.9	1900.5 ± 475.0	-31.52 ± 0.80	12.04 ± 0.33	0.51 ± 0.07
Mahony	20	12.4 ± 1.8	671.5 ± 153.1	4166.5 ± 3001.4	-26.06 ± 0.94	10.62 ± 0.76	0.37 ± 0.28
Kelly	20	12.3 ± 4.5	592.8 ± 82.9	2522.0 ± 1177.8	-30.41 ± 0.99	12.47 ± 0.40	0.47 ± 0.19
Mirror	20	-	468.9 ± 41.9	1239.0 ± 287.9	-29.83 ± 1.52	9.66 ± 0.58	0.68 ± 0.29
Ste. Thérèse	11	24.9 ± 5.2	708.1 ± 60.7	3217.9 ± 888.0	-28.17 ± 0.67	13.49 ± 0.82	0.77 ± 0.47
Stark	25	23.2 ± 4.6	726.1 ± 74.3	4559.2 ± 1989.8	-26.29 ± 2.42	12.57 ± 0.72	0.47 ± 0.40
Willow	32	15.8 ± 4.8	613.7 ± 92.9	3067.5 ± 2159.8	-27.70 ± 1.08	13.07 ± 0.63	0.38 ± 0.08
Great Slave	46	13.4 ± 6.1	584.9 ± 83.4	2693.0 ± 1557.8	-28.09 ± 1.91	12.39 ± 0.66	0.17 ± 0.07
Cli	25	16.9 ± 7.2	499.6 ± 140.1	1651.2 ± 1792.0	-26.53 ± 1.18	11.65 ± 1.25	0.92 ± 1.08
Little Doctor	10	-	547.3 ± 42.3	1603.0 ± 350.0	-29.91 ± 0.91	12.34 ± 0.50	0.39 ± 0.08
Trout	28	16.7 ± 5.2	650.5 ± 40.6	2667.6 ± 734.4	-27.34 ± 0.97	12.14 ± 0.57	0.39 ± 0.07
Mean	**24.9**	**15.4 ± 4.4**	**597.3 ± 81.6**	**2582.6± 1028.8**	**-27.64 ± 2.11**	**12.16 ± 1.45**	**0.43 ± 0.20**

Carbon isotope values ranged from -31.5‰ in Turton Lake to -24.1‰ in Aubry Lake, with a mean and standard deviation of -27.83 ± 1.4‰. Nitrogen isotope values ranged from 9.7‰ in Lac Ste. Thérèse to 16.2‰ in Colville Lake with a mean and standard deviation of 12.2 ± 0.8‰. However, $\delta^{15}N$ values in both Lac Ste. Thérèse and Mirror Lake were anomalous compared with those in the other study lakes, and when removed, the mean values ranged only from 10.6 to 13.5‰.

Mean THg concentrations in lake trout muscle tissue ranged from 0.17 µg Hg/g in Great Slave Lake to 0.92 µg Hg/g in Cli Lake, with an overall across-lake mean and standard deviation of 0.43 ± 0.2 µg Hg/g. Four of the study lakes, Cli, Ste. Thérèse, Mirror, and Turton, had mean THg concentrations in lake trout muscle that were above the 0.5 µg Hg/g Canadian Food Inspection Agency (CFIA) guidelines for the CSFG. All of the study lakes except for Great Slave Lake, the only one supporting a commercial fishery, contained lake trout populations with mean mercury concentrations that were above the 0.2 µg Hg/g guidelines set by Health Canada for the frequent consumers of fish (FCFG).

2.4.2. Physical characteristics of study lakes

The lakes range in size from 2.96 km^2 (Mirror Lake) to 31,328 km^2 (Great Bear Lake) with a wide range of sizes between the two extremes. The watershed area/lake surface area ratio is a measure of the ratio of the area of the immediate drainage basin surrounding that lake to that of the total lake surface area. These values ranged from 2.0 (Aubry Lake) to 34.5 (Great Slave Lake) (Table 2.1).

All of the lakes in this study were cold, slightly alkaline lakes that were well oxygenated. Temperature steadily fell from the surface to the bottom in all lakes as is typically observed in summer. None of the lakes were stratified at the time of sampling. The DO concentrations ranged from 7.9 mg/L in Willow Lake to 13.4 mg/L in Great Bear Lake. The pH ranged from 7.9 to 8.7 (Table 2.3). Conductivity ranged from 28 µS/cm in Stark Lake, which is on the Canadian Shield, to 842 µS/cm in Kelly Lake, located on glacial till. Turton and Kelly lakes had very high conductivity values, while all other lakes were more dilute with conductivities of less than 300 µS/cm. Median values of field parameters are reported in Table 2.3.

All the lakes had low TP and Chl*a* concentrations suggesting low productivity. Total phosphorous concentrations ranged from 0.003 to 0.025 mg/L.

Table 2.3. Lake characteristics including pH, conductivity, total phosphorus (TP), chlorophyll *a* (Chl*a*), dissolved organic carbon (DOC), dissolved oxygen (DO), temperature, total mercury (THg) in water, methylmercury (MeHg) in water, and mercury in zooplankton measured in all 17 study lakes.

Lake	Sampling Date	Temperature * (°C)	pH *	Conductivity * (µS/cm)	DO* (mg/L)	TP** (mg/L)	Chl. *a*** (µg/L)	DOC** *(mg/L)	THg in water** * (ng/L)	MeHg in water** (ng/L)	Hg in Zooplankton (µg/g)
Aubry	-	-	-	-	-	-	-	-	-	-	-
Colville	August 2002	11.6	8.1	170	11.0	0.016	4.53	5.3	1.47	0.06	0.005
Manuel	Summer 1999	16.6	8.7	195	9.1	0.007	-	-	-	-	-
Rorey	Summer 1999	16.9	8.1	221	8.8	0.005	-	-	-	-	-
Belot	Summer 1999	13.4	8.5	168	9.8	0.003	-	-	-	-	-
Great Bear	August 2002	4.79	8.0	152	13.4	0.006	1.18	1.6	1.50	0.03	0.003
Turton	Summer 1999	18.6	8.6	518	8.3	0.007	-	-	-	-	-
Mahony	Summer 1999	-	-	-	-	0.025	-	-	-	-	-
Kelly	August 2002	12.3	8.4	843	10.8	0.006	1.53	4.2	1.66	0.03	0.008
Mirror	August 2002	9.0	8.2	230	12.3	0.007	1.78	6.2	1.90	0.02	0.010
Ste. Thérèse	August 2002	14.2	8.1	243	9.7	0.010	1.78	10.8	3.13	0.15	0.009
Stark	August 2003	-	-	28	-	0.006	2.41	4.3	0.88	0.00	0.006
Willow	July 2001	15.7	8.6	154	7.9	0.012	1.56	10.0	0.11	0.04	0.004
Great Slave	July 2001	14.8	8.0	200	9.5	-	1.2	-	-	-	0.003
Cli	August 2002	7.6	8.0	202	11.1	0.010	0.14	7.0	1.23	0.04	0.005
Little Doctor	August 2002	10.6	8.9	154	11.1	0.011	1.42	14.7	1.95	0.07	0.004
Trout	August 2003	14.8	7.9	137	8.7	0.010	6.92	12.5	1.01	0.02	0.003

* - profile median

** - concentrations in surface water grab sample

All TP concentrations were < 0.010 mg/L except for Colville (0.016 mg/L), Mahony (0.025 mg/L), Willow (0.012 mg/L), and Little Doctor (0.011 mg/L) lakes. Chlorophyll *a* concentrations were low, ranging from 0.14 to 6.92 µg/L. Only Colville, Stark, and Trout lakes had Chl*a* concentrations above 2.0 µg/L (Table 2.3). Dissolved organic carbon concentrations ranged from 4.2 mg/L in Kelly and Stark lakes, to 14.7 mg/L in Little Doctor Lake.

Total mercury and MeHg concentrations in water were low to undetectable in all lakes. Total mercury concentration ranged from 0.9 to 3.1 ng Hg/L. Methylmercury concentrations in water ranged from undetectable (<0.02 ng Hg/L) to 0.15 ng Hg/L. Total mercury and MeHg concentrations were below the Canadian water quality guidelines for protection of aquatic life set for mercury, i.e., 26 ng Hg/L for THg in water and 4 ng Hg/L for MeHg in water (CCME 1999).

2.4.3. Within-lake relationships

Total mercury (log transformed) concentrations in lake trout in the 17 study lakes were significantly ($p < 0.05$) correlated with fish age, fork length, weight, $\delta^{13}C$, and $\delta^{15}N$ although the nature of these relationships varied with each lake (Table 2.4). All correlations were positive with the exception of $\delta^{13}C$. Total mercury concentrations were significantly correlated with age in only four of 15 lakes (fish age could not be determined in Little Doctor or Mirror lakes presumably because of the very old age of the fish, which is associated with calcification of the otolith); these lakes were Cli, Great Bear, Aubry, and Great Slave. Log mercury concentrations were significantly correlated with fork length in 12 of the 17 lakes while log mercury and weight was significantly correlated in only nine lakes. The strongest correlations were for Aubry, Cli, and Little Doctor lakes. Trout, Willow, Mirror, and Rorey lakes showed no significant relationships between any of the measured variables and tissue mercury concentration. When feeding relationships were explored, log mercury was significantly correlated with $\delta^{13}C$ in only five of the 17 lakes and with $\delta^{15}N$ in nine lakes.

Overall, fork length was the best predictor of mercury concentrations in lake trout followed by weight and $\delta^{15}N$. The value of $\delta^{15}N$ as a predictor is somewhat constrained by the fact that acquiring the data involves a chemical measurement costing about the same as a mercury measurement, although its value as an explanatory variable cannot be discounted.

Table 2.4. Pearson product moment correlation coefficients (r) for within-lake correlations between age, fork length, weight carbon and nitrogen stable isotopes and log total mercury tissue concentrations in lake trout from the 17 study lakes in the Northwest Territories.

Lake	n	Age	Fork Length	Weight	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
Aubry	20	0.74*	0.79*	0.74*	-0.04	0.66*
Colville	39	0.24	0.59*	0.52*	-0.32	0.23*
Manuel	19	0.57	0.53*	0.25	-0.08	0.33
Rorey	20	0.04	0.03	0.00	-0.36	0.30
Belot	20	0.00	0.61*	0.47*	-0.73*	0.74*
Great Bear	10	0.86*	0.74*	0.62	-0.43	0.89*
Turton	20	0.31	0.68*	0.51*	-0.20	0.00
Mahony	20	0.60	0.93*	0.88*	-0.52*	0.69*
Kelly	20	0.45	0.47*	0.67*	-0.23	0.08
Mirror	20	CA[a]	0.03	0.07	-0.36	0.15
Ste. Thérèse	11	0.52	0.23	0.44	-0.30	0.92*
Stark	25	0.14	0.41*	0.30	-0.49*	0.80*
Willow	32	0.14	0.00	0.03	-0.05	0.34
Great Slave	46	0.40*	0.54*	0.51*	-0.37*	0.40*
Cli	25	0.88*	0.88*	0.83*	-0.78*	0.61*
Little Doctor	10	CA[a]	0.77*	0.69*	-0.07	0.26
Trout	28	0.16	0.06	0.26	-0.14	0.07

* Relationships are significant at $p < 0.05$.

[a] Ages could not be determined for lake trout in these lakes.

Table 2.5. Pearson product moment correlation coefficients (r) for correlations between measured variables pooled from all 17 study lakes in the Northwest Territories and log total mercury concentration in lake trout tissue. Graphs of significant relationships are shown in Appendix B.

Variable	n	r value	*p* value
Age	256	0.479*	<0.001
Fork length	365	0.201*	<0.001
Weight	365	0.162*	<0.001
$\delta^{13}C$	365	-0.180*	<0.001
$\delta^{15}N$	365	0.007	0.89
Latitude	365	-0.099	0.06
Log lake area	365	-0.560*	<0.001
Watershed / area ratio	365	-0.294*	<0.001
Total mercury in zooplankton	246	0.476*	<0.001
pH	301	-0.090	0.77
Dissolved organic carbon	200	0.037	0.61
Total phosphorus	299	-0.166	0.05
Conductivity	325	0.197	0.50
Temperature	301	0.054	0.37
Chlorophyll *a*	246	-0.098	0.13
Dissolved oxygen	301	0.087	0.13
Total mercury in water	200	0.169*	0.02
Methylmercury in water	200	0.039	0.59

* Relationships are significant at $p < 0.05$.

2.4.4. Among lake relationships

Linear regression analyses were performed to investigate significant relationships individually between log THg tissue concentration in lake trout and all biological, chemical, and physical variables and pooled together from all lakes. Pearson product moment correlation coefficients and corresponding *p*-values are listed in Table 2.5, and graphs of significant relationships are shown in Appendix B.

Considering first the biological features, there was a statistically significant ($p < 0.05$) positive correlation between log THg tissue concentration and each of fish age, fork length, and weight and a negative correlation with $\delta^{13}C$ ($\delta^{15}N$ was not statistically significant). Considering geographic features) log total lake area and the watershed to lake surface area ratio were statistically significant ($p < 0.05$) with negative correlations: latitude was significant at $p = 0.06$). Among the chemical variables, only mercury in zooplankton was statistically significant at $p < 0.05$ and total phosphorus at $p = 0.05$). Temperature, pH, and conductivity were not statistically significant.

2.4.5. Multiple regression

Best subsets regression analyses were performed to determine the best fitting regression model for predicting THg concentration in a given lake trout based on stock assessment and other simple studies and considerations. Physical variables considered were log lake surface area, the ratio of watershed area to lake surface area and latitude. Latitude was considered because the correlation between log mercury level in lake trout and latitude had a *p*-value of 0.05, suggesting that this variable could be significant. The biological variables considered were lake trout fork length and age. Weight was excluded because it introduced multicollinearity to the model. Carbon^{13} stable isotope and $\delta^{15}N$ were not included because these variables are not traditionally measured in stock assessment studies and mercury could be measured just as easily. The mercury concentration in zooplankton and THg concentration in water were excluded from the model because they are not measured during stock assessment studies; had these variables been included, several lakes would need to have been excluded from the model development.

Log THg tissue concentration in any given lake trout can be described by a regression (equation 2.2) based on two biological features of the fish (fish length and age) and three physical features of the lake it inhabits (log surface area, watershed

area to lake surface area ratio and latitude). This model explains 73% of the variance, and the Mallows c_p value was a conservative 6.0 (which is equal to p+1). The resulting model was:

$$\log Hg = 0.698 - (0.0156 \times latitude) + (0.0031 \times age) + (0.000535 \times length) - (0.245 \times \log lake\ area) + (0.00675 \times watershed\ area/lake\ area\ ratio),\ r^2 = 0.73 \quad (2.2)$$

Model 1 includes lake trout age, which is not a variable readily obtained by untrained personnel. Therefore, a second regression (equation two) was calculated with age removed from the model. This regression explained 71% of the variance. Model 2 was:

$$\log Hg = 0.724 - (0.0174 \times latitude) + (0.000588 \times length) - (0.2193 \times \log lake\ area) + (0.00197 \times watershed\ area/lake\ area\ ratio),\ r^2 = 0.71 \quad (2.3)$$

The relationship between measured and predicted mercury concentrations is shown for the two models in Figure 2.2. As expected, there was a strong positive correlation between measured and predicted mercury concentrations with relatively narrow 95% confidence intervals.

Predictive model 1 had the following equation:

$$\log measured\ Hg = 1.05 \times \log predicted\ Hg + 0.01,\ r^2 = 0.73 \quad (2.4)$$

Predictive model 2 had the following equation:

$$\log measured\ Hg = 1.11 \times \log predicted\ Hg + 0.06,\ r^2 = 0.71 \quad (2.5)$$

Measured and predicted mercury concentrations were graphed individually for each of the 17 lakes (Figure 2.3). There was an excellent relationship between predicted and measured mercury concentrations for most lakes. Cli Lake, where the lake trout were small but very old, showed the poorest relationship; mercury concentrations were higher than predicted. Also lake trout from Great Bear Lake, which is a large and very low oligotrophic lake, had mercury concentrations lower than predicted. Including age as a factor in the model did little to improve the relationship between measured and predicted mercury concentrations in most lakes. With the exception of Cli Lake, the models were good at predicting which lakes contained lake trout that would exceed the 0.5 µg/g guideline for the commercial sale of fish (CSFG) and with the exception of Great Bear Lake, fish which would exceed the 0.2 µg/g guideline for FCFG.

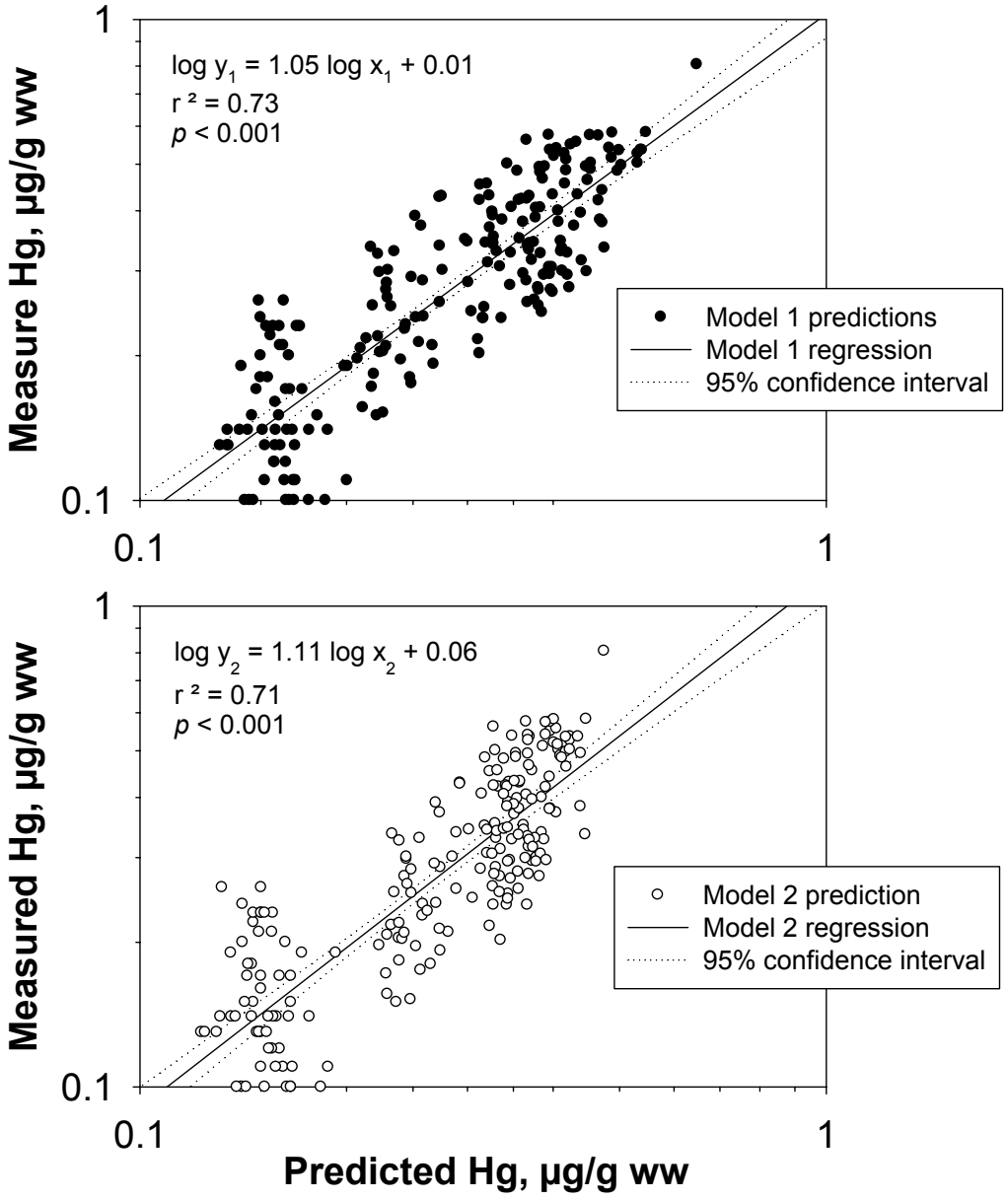


Figure 2.2. Regression of measured total mercury concentration in lake trout dorsal muscle tissue versus predicted total mercury concentration in muscle tissue derived from multiple regression equations. The upper panel graph is based on Model 1 which includes latitude, age, fork length, log of lake surface area, and watershed area to lake area ratio as predictor variables. The lower panel graph is based on Model 2 which includes latitude, fork length, log of lake surface area, and watershed area to lake area ratio as predictor variables.

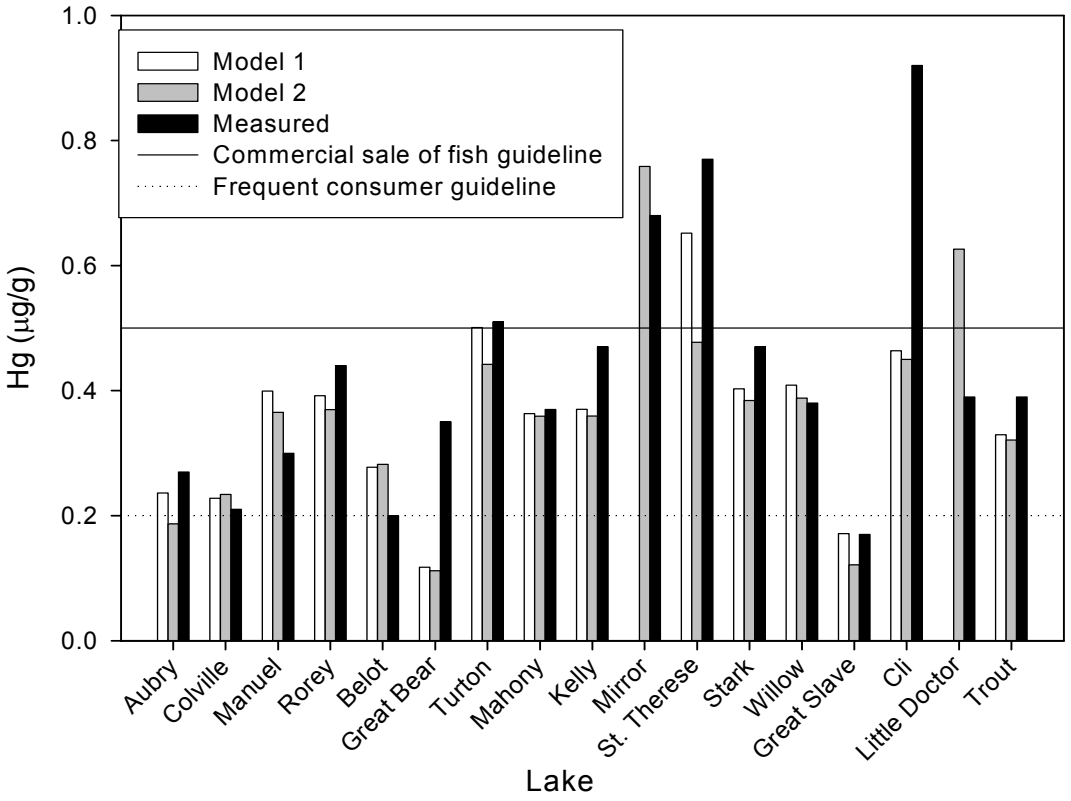


Figure 2.3. Measured and predicted mercury concentrations in lake trout dorsal tissue from all 17 study lakes in the Northwest Territories. Model 1 corresponds to predictions made using Equation 2.2, using latitude, fork length, age, lake area and watershed to lake area ratio as predictors. Model 2 corresponds predictions made with Equation 2.4, which uses the same variables as Model 1 except age, which is excluded.

2.4.6. Model accuracy

The accuracy of the models was explored using data from two other lakes in the NT, Canada, Rae and Faber lakes. Lake trout averaged 15.1 years in Rae Lake and 14.7 years in Faber Lake while mean fork length was 553.6 mm and 538.9 mm respectively; lake trout were slightly younger and smaller than those from the 17 study lakes (Table 2.6). The lakes are moderately large (252.5 and 439 km^2 respectively), and among the larger lakes (excluding Great Bear and Great Slave Lake) in the MRB study. The watershed area to lake surface area ratio also was low (3.4 - 3.7). Mean mercury concentration averaged 0.46 ± 0.14 μg/g in Rae Lake lake trout and 0.35 ± 0.07 μg/g in 0.07 in Faber lake fish. The predicted concentration in Rae Lake lake trout was 0.30 μg/g and in Faber lake fish was 0.25 μg/g (Figure 2.4). One-way ANOVA shows no significant difference between the values predicted by equations 1 and 2, however, both values were significantly lower than the actual mercury concentration measured in fish from both lakes. The models did correctly predict that the THg concentrations in lake trout from Rae Lake were higher than those in Faber Lake. It also accurately predicted that mean mercury concentrations in lake trout would be >0.2 μg/g but less than 0.5 μg/g.

2.5. Discussion

Mercury concentrations in piscivorous fish species in some lakes in the MRB, are commonly found to have tissue THg concentrations exceeding the 0.5 μg Hg/g CFIA guideline for CSFG. Total mercury concentrations in lake trout tissue exceed the 0.2 μg Hg/g Health Canada guideline set for FCFG in a surprising 94% of the lake trout sampled. Possible explanations for these high THg concentrations are many and varied. Inorganic mercury transformation to MeHg, and subsequent accumulation in biological tissues, is affected by many different variables in the aquatic environment. This makes the study of mercury contamination a regional problem; on a global scale solutions cannot be determined and applied to all lakes affected by high mercury concentrations. Rather, each region that is determined to be contaminated must be studied to determine the variables influencing high mercury concentrations.

Table 2.6. Variables associated with Rae and Faber lakes and their respective lake trout populations including mean age, fork length, latitude, and lake and watershed area relative to lake area, that are used in predictive models 1 and 2.

Lake	Latitude	Mean age (years)	Mean fork Length (mm)	Surface Area (km^2)	Watershed area / lake area
Rae	64° 10' N	15.1 ± 5.0	553.6 ± 35.4	251.5	3.35
Faber	63° 56' N	14.7 ± 5.2	538.9 ± 59.1	439.0	3.66

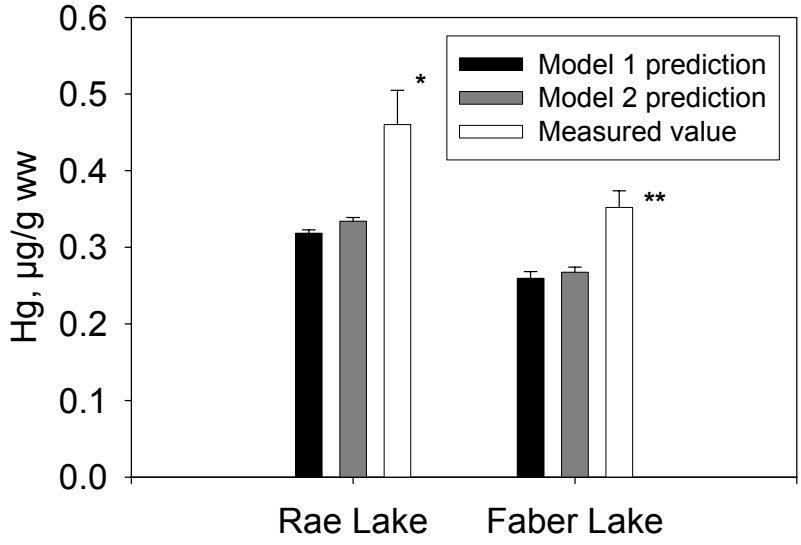


Figure 2.4. Mean total mercury concentrations predicted using models 1 and 2, and measured in lake trout in Rae and Faber lakes, NT, Canada. * significantly different from the estimates created by both models 1 and 2, $p = 0.001$. ** significantly different from the estimates created by both models 1 and 2, $p < 0.001$. Error bars show standard error from the mean.

Mean fish age in the NT study lakes was 15.2 years; very old when compared to other populations studied in the past including northern Alberta and Saskatchewan (NAS) lakes where the lake trout had a mean age of eight years (Muir *et al.* 2001). The higher mean age of lake trout in the NT lakes is believed to result from the negligible fishing pressures due to the region's low human population, modest sport fisheries, and no commercial fisheries with the exception of Great Slave Lake (Evans *et al.* 2005). MacCrimmon *et al.* (1983) and Evans *et al.* (2005) determined that age was significantly and positively correlated to lake trout THg tissue concentration. Lange *et al.* (1993) found the same correlation in large mouth bass.

The present study found that age was related to mercury concentration in only four of the 15 lakes where fish age was determined. This may be due to the narrow range in ages of the fish caught in the study. Age bias in the study could be due to nets used and/or to the fact that recruitment is limited by a relatively large population of large adults that maintain younger lake trout at low levels. Fish age is significant because as lake trout get older and larger, they alter their diet from primarily invertebrates to primarily forage fish species, when such prey are available (MacCrimmon *et al.* 1983; Vander Zanden and Rasmussen 2001). This diet shift is generally accompanied by an increase in mercury concentration in fish tissues. When all lakes were considered and data pooled, lake trout age was significantly correlated with total mercury tissue concentration in lake trout (Pearson correlation r value = 0.48, $p < 0.001$). The greater correlation between THg and age in the pooled data than in individual lakes could be due to a larger data set resulting in a greater range of ages, allowing more of a relationship to appear.

Fish fork length and weight are often correlated with contaminant concentration. Both Rose *et al.* (1999) and Lange *et al.* (1993) found that fish weight was a strong predictor in Florida lakes. In this study, total mercury tissue concentration in lake trout was positively correlated with mean lake trout fork length and weight in 11 and nine of the 17 lakes, respectively. It is possible that the lack of a relationship in the remaining lakes is due to the presence of different genetic morphs (phenotypic varieties in a polymorphic population) of lake trout. Howland *et al.* (2004) has found at least five different morphs of lake trout in Great Bear Lake. The different morphs appear slightly different with respect to fin size, length/weight relationships, jaw shape, and diet. All of these variables may affect mercury concentration in lake trout. However, when data from all lakes were pooled together

both fork length and weight were significantly correlated with total mercury concentration in lake trout tissue (r = 0.20, 0.16, $p < 0.001$, < 0.001, respectively, Table 2.5).

Total mercury tissue concentration in lake trout from the study lakes seemed to be little affected by their feeding habits, as measured through stable isotope (SI). Total mercury tissue concentration was significantly correlated with $\delta^{15}C$ in only four of the 17 lakes, and $\delta^{15}N$ in nine of the lakes. However, when all lakes were pooled, $\delta^{15}C$ were significantly and negatively correlated with THg concentration in lake trout tissue (r = -0.18, $p < 0.001$). Low $\delta^{15}C$ is often associated with more pelagic food webs, which are correlated with higher tissue mercury concentrations in fish. Power *et al.* (2002) noted a similar trend. There was no relationship between THg concentration in lake trout tissue and $\delta^{15}N$ when the lakes were pooled. Low $\delta^{15}N$ in Mirror Lake and Colville Lake may have been the primary factors contributing to this lack of a significant relationship between total mercury and $\delta^{15}N$. Furthermore, the inclusion of younger fish (<6 years) in the study might have lead to a clearer relationship between $\delta^{15}N$ and THg tissue concentration in lake trout because of a probably increase in the range of $\delta^{15}N$ values. Young fish are more insectivorous, while older lake trout feed at higher trophic levels, consuming mainly forage fish species.

As Bodaly *et al.* (1993) and Håkanson (1991) also determined, THg tissue concentration in lake trout was significantly and negatively correlated with lake surface area (r = -0.56, $p < 0.001$, Table 2.5). Small lakes are more conducive to mercury transformation from inorganic to MeHg because of higher mean water temperatures. Warm water temperatures lead to increased fish metabolism, increasing both feeding and respiration, both of which lead to increased mercury uptake. These small lakes are also more affected by watershed inputs. There was a weak, but significant, positive correlation between total mercury concentration in lake trout tissue and watershed area to lake surface area ratio (r = 0.29; $p < 0.001$, Table 2.5). Other studies have also found watershed inputs to influence mercury concentrations (Bodaly *et al.* 1993; Suns and Hitchin 1990; Rose *et al.* 1999). Increased watershed area relative to lake surface area leads to increased inputs of both inorganic mercury and MeHg on a per volume basis from the surrounding environment.

Total mercury tissue concentration was positively correlated with total mercury concentration in lake water. However, THg and MeHg concentration in water were only measured in ten of the 17 lakes, so including this variable in a multiple regression would have weakened the predictive capability of the model by reducing the number of observations.

While research in other regions has shown correlations between mercury concentration in fish tissue and various water quality paramaters such as pH, this study found no such relationships. The lakes located in the MRB are all moderately alkaline, ranging in pH from 8.0 to 8.6. This is in contrast to other studies which found that higher mercury concentrations in aquatic organisms are associated with acidic pH; in these studies pH ranged from > 6 to < 8 (Grieb *et al.* 1990; Miskimmin *et al.* 1992; Lange *et al.* 1993; Schuehammer and Graham 1999). Greenfield *et al.* (2001) did investigate pH and mercury concentrations in yellow perch, (*Perca flavescensI*), from circumneutral lakes, and found a weak positive relationship; however, the pH range of their study lakes ranged from 7.0 to 8.8, a larger range with more neutral values than in this study.

Total mercury tissue concentration was positively correlated ($r = 0.476$, $p < 0.001$; Table 2.5) with mercury concentration in zooplankton, which are at the base of the lake trout food chain. However, THg in zooplankton was only measured in 11 of the 17 lakes, and data from the most northern lakes is noticeably absent. Concentrations of mercury in zooplankton are important because zooplankton are at the base of the food chain level, where mercury moves from water into aquatic organisms, and begins biomagnifying. Lake trout acquire more mercury from food intake than from water intake during respiration (Hall *et al.* 1997). This correlation has been found in other studies (France 1995; Plourde *et al.* 1997; Muir *et al.* 2001; Campbell *et al.* 2003; Kidd *et al.* 2003) and will be further investigated in the next chapter.

2.5.1. Multiple regression

Most of the variation in lake trout mercury tissue concentration could be explained by five variables: fish age, fish fork length, latitude, log lake area, and watershed area to lake surface area ratio. These factors were used to develop a predictive model of mercury concentrations in lake trout. The regression equation using these five variables explained 73% of the variation in THg concentration in

lake trout tissue. This model could readily be used in stock assessment studies where mercury concentrations in fish were not measured. However, age determination requires specialized analyses and cannot be performed on all lake trout populations. Therefore, the model was further simplified by removing age. Without fish age as a variable, the model explained 71% of the variance.

Both models generally serve well as predictors of mercury concentrations in lake trout in the 17 study lakes. Both models overestimated mercury concentrations in Cli Lake, where the trout are small but very old, and underestimated mercury concentrations in Great Bear Lake, which is a very large low productivity lake with old lake trout. However, with these exceptions, the models correctly predicted the lakes in which lake trout would exceed the 0.5 μg Hg/g CSFG and the 0.2 μg Hg/g FCFG. When the models were used to predict THg tissue concentrations in lake trout from Rae and Faber lakes, the predicted concentrations were somewhat lower than the measured concentrations. However, both lakes are on the Canadian Shield, and in a different geological setting for which the models were developed.

2.6. Conclusions

Elevated mercury concentrations in lake trout in many lakes in the NT are most often associated with large, old fish in lakes with large watersheds and inversely correlated with small lakes and latitude. These variables together can be used in regression models to explain more than 73% of the variance in mercury concentrations in lake trout over the 17 study lakes. As a screening tool, the model is useful in assessing whether or not lake trout inhabiting a particular lake are likely to have mean mercury levels which exceed 0.2 μg Hg/g FCFG and 0.5 μg Hg/g CSFG.

3.0 COMPARISON OF TROPHIC RELATIONSHIPS IN AQUATIC FOOD WEBS IN TWO SERIES OF LAKES IN NORTHERN CANADA, AND RESULTING EFFECTS ON MERCURY CONTAMINATION IN LAKE TROUT

3.1. Abstract

Differences in feeding and food web biomagnification rates can translate into differences in contaminant accumulation by predatory species. In this study, two groups of lake trout (*Salvelinus namaycush*) one from a series of lakes in the Northwest Territories (NT), and the other from a series of lakes in northern Alberta and Saskatchewan (NAS), were compared to determine whether differences in food web biomagnification rates and trophic feeding could account for differences in total mercury concentration in muscle tissue. Lake trout from the NT lakes had significantly higher mercury concentrations than those from the NAS lakes ($p < 0.001$). However, despite the difference in mercury concentration, lake trout from the two areas fed at similar trophic levels. Therefore, feeding habits did not explain the difference in tissue mercury concentration. The biomagnification rate of mercury (defined as the relationship between nitrogen isotope ($\delta^{15}N$) vs. log total mercury (THg) concentration) differed between the two study areas, i.e., mercury biomagnification occurred at a slower rate in NT than in the NAS lakes. However, because the mean age of the lake trout from the NT lakes was greater than in NAS lakes, lake trout from the NT lakes accumulated mercury over a longer period of time, ultimately leading to higher total mercury concentrations in their muscle tissue. Northwest Territory food webs had more negative carbon isotope ($\delta^{13}C$) values inferring that zooplankton, aquatic insects and fish relied more on pelagic than littoral zone carbon sources. Other studies have shown that more pelagic food webs have been associated with higher mercury concentrations.

3.2. Introduction

Research studies conducted throughout the last decade uncovered higher than expected levels of mercury in muscle tissue of fish from remote north-western

Canadian lakes (Shilts and Coker 1995; Stephens 1995; Lockhart *et al.* 2005). The water in many of these lakes has very low to undetectable mercury concentrations (Chapter 2; Evans *et al.* 2005), leading researchers to believe that the high total mercury (THg) concentrations in fish tissue were likely due to factors related to increased bioaccumulation and biomagnification within the food chain rather than uptake from a mercury-rich environment. In Chapter 2, it was shown that fish age, fork length, latitude, watershed area to lake surface area ratio all played an important role in the accumulation of mercury in lake trout (*Salvelinus namaycush*) tissue in a series of 17 study lakes in the Northwest Territories (NT).

Lake trout living south of the NT study area could be expected to have higher mercury concentrations for two reasons. First, lake trout living in areas such as northern Alberta and Saskatchewan (NAS) could be expected to have higher mercury levels because of their greater proximity to anthropogenic sources. Second, mercury levels in the NT were inversely related to latitude, possibly reflecting the shorter growing season, lower water temperatures and lower productivity in the more northern lakes. However, THg concentrations in the NT lake trout were, on average, higher than in the NAS lake trout although similar sized lakes and fish were studied in both of the areas (Evans *et al.* 2005). Lake trout are known to feed on a variety of different food sources based on availability, and, if there are differences in the feeding habits of NT and NAS lake trout, this could account for some of the differences in mercury levels.

Diet is an important determinant of contaminant levels in fish because methylmercury (MeHg) biomagnifies through food webs (Verta 1990; Bodaly *et al.* 1993). Piscivorous fish have higher tissue concentrations of mercury than planktivorous or omnivorous species (Rasmussen *et al.* 1990). Lake trout are facultative predators (Scott and Crossman 1998). While they prefer to feed on forage fish species, when abundant and available, they can survive on zooplankton and invertebrates. Therefore, differences in food sources could help explain differences in THg concentrations in lake trout.

Previously researchers relied on stomach content analyses to determine major food sources. However, this approach is time consuming and provides only an instantaneous view of an individual's feeding habits. More recently, stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope analysis techniques were developed allowing for time-integrated data on the diet and relative trophic position of fish populations.

Stable nitrogen isotopes analysis provides a measure of the dietary nitrogen accumulated in the tissues over the period of tissue turnover, which can encompass months to years (Hesslein *et al.* 1993). Nitrogen isotopes consistently fractionate in organisms; ^{15}N is retained in tissues, while ^{14}N is eliminated. Therefore, with each trophic transfer $\delta^{15}N$ values increase making stable nitrogen isotopes useful in determining the relative trophic position of biota. The average difference in $\delta^{15}N$ signatures (ratio of $^{15}N/^{14}N$) between trophic levels is approximately 3 to 4 ‰ (DeNiro and Epstein 1978; Vander Zanden and Rasmussen 2001). The consistency of nitrogen enrichment at each trophic level provides a quantitative measure of the relative trophic position of an organism within a food web (Cabana and Rasmussen 1994), which can then be correlated with contaminant analysis to estimate metal uptake and biomagnification rates (Power *et al.* 2002). Once relative trophic position and contaminant concentration is determined for a range of food types (e.g., zooplankton, aquatic insects, forage small fish, predatory fish) contaminant bioaccumulation can be investigated through regression analyses of contaminant concentrations versus $\delta^{15}N$ (Cabana and Rasmussen 1994). This type of analysis has been useful in investigating mercury biomagnification freshwater systems (Kidd *et al.* 1995; Gorski *et al.* 2003).

Carbon signatures (ratios of $^{13}C/^{12}C$) are largely unaffected by trophic level, and undergo approximately 1‰ or less enrichment in $\delta^{13}C$ per trophic level. Pelagic and benthic algae in freshwater exhibit distinct carbon signatures due to different fractionation during carbon fixation (Hecky and Hesslein 1995). Free-floating pelagic algae close to the water surface can access a large dissolved CO_2 reservoir, which fractionate only slightly, resulting in lighter $^{13}C/^{12}C$ ratios of approximately -29 ‰. At the sediment/water interface there is a more limited CO_2 reservoir available and carbon-limited benthic algae are less isotopically discriminating, resulting in heavier $\delta^{13}C$ carbon signatures between -25 and -10 ‰ (Hecky and Hesslein 1995; Bootsma *et al.* 1996). These differences are maintained and passed up the food chain, indicating the origin of organic carbon in organisms at upper trophic levels (Hecky and Hesslein 1995; France 1995). Because there is little change in the carbon signature through successive trophic levels, the carbon signature can be used to differentiate between the relative reliance of organisms on food webs of pelagic and littoral origin. These differences can help determine the relative importance of near-shore versus pelagic sources in an organism's diet

(Bootsma *et al.* 1996). Measurements of both nitrogen and carbon isotopes, when used together, can provide a detailed understanding of lacustrine community food webs. Also, when coupled with contaminant analysis, stable isotope analysis can be used to trace the pattern and extent of biomagnification of contaminants in aquatic food webs.

Linear regressions of log transformed THg concentrations and $\delta^{15}N$ can be calculated with the resulting slope providing a numerical estimate of the rate of biomagnification of a contaminant. These calculated slopes can also provide a means of comparing food chains from different lakes and regions. When the slopes are similar, biomagnification is occurring at a similar rate. A steeper slope indicates that the contaminant is biomagnifying at a faster rate. The y-intercept of the regression is influenced by the bioavailable concentration of a contaminant in the environment as well as the baseline $\delta^{15}N$ value in that food web. A higher intercept suggests greater concentrations of the contaminant at the base of the food web and in the water.

Differences in biomagnification rates may help to explain the difference in mercury concentration in lake trout between the NT and NAS study areas, i.e., the higher THg concentrations in NT than NAS lake trout could occur if the food webs in the NT lakes are associated with steeper biomagnification slopes. In addition, a higher y-intercept, in NT than NAS lakes could explain some of these regional differences; however, a lower y-intercept is expected for the NT lake food webs because lakes in the far north are further from anthropogenic sources of pollution.

The first objective of this thesis chapter was to determine the trophic relationships of lake trout in a series of lakes in the NT (i.e., to determine if they were feeding primarily on invertebrates, omnivores, piscivores, or even as cannibals). Second, these data were compared with similar $\delta^{15}N$ vs. log THg tissue relationships for lake trout from the NAS lakes to assess whether mercury biomagnified different rates. Third, the intercepts of the biomagnification rate regressions were compared to assess whether mercury was more bioavailable at the base of the food web in NT than NAS lakes. Finally, pelagic versus littoral zone feeding and fish age were investigated as potential factors affecting mercury concentrations in lake trout in the two regions.

3.3 Materials and Methods

3.3.1. Study areas

The lakes in the NT study area are located in the Mackenzie River Basin (MRB), along both sides of the Mackenzie River from Trout Lake in the south to Aubry Lake in the north. Great Slave Lake was not included in the analysis (as in Chapter 2) because there was no data available on mercury concentrations in forage fish and aquatic invertebrates.

The topography and geology of this region varies greatly, including the Mackenzie and Franklin mountains as well as the Horn Plateau and numerous other isolated hilly areas. Stark and Great Slave lakes lie partly on the Canadian Shield while all the other lakes lie on palaeozoic deposits. The climate is sub arctic with mean annual temperatures averaging from -5.4 to -8.7°C (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/results_e.html, accessed January 18, 2005). The study lakes vary widely in total surface area from 3 km^2 (Mirror Lake) to 31,328 km^2 (Great Bear Lake) All lakes are located in a northern boreal ecosystem characterized by tree species including white spruce, (*Picea glauca)*, birch, (*Betula papyrifera var. papyrifera)*, trembling aspen, (*Populus tremuloides)*, and jack pine, (*Pinus banksiana)*. In low-lying areas the trees give way to muskeg. The lakes are oligotrophic to mesotrophic, meaning that they are low to moderate in productivity, with total phosphorus (TP) concentrations of 0.003 to 0.016 mg/L, and chlorophyll a (Chl*a*) concentrations of 0.14 to 4.53 µg/L (Chapter 2). The lakes are highly-oxygenated drainage lakes, with summer dissolved oxygen (DO) concentrations ranging from 7.9 to 13.4 mg/L (Chapter 2). They are covered by ice from October until mid-June (Figure 3.1(a), Table 3.1(a)).

The NAS lakes are located within the boreal northlands ecosystem. High plateaus and hills characterize this region, and the dominant tree species include trembling aspen and white spruce (Mitchell and Prepas 1990). Mean annual temperatures range from 0 to 2.1°C (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/index_e.html, accessed January 18, 2005). Lakes in NAS are in closer proximity to anthropogenic activities than those studied in the NT, possibly leading to higher contaminant loading. Many of the NAS lakes have been and are currently affected by mining, particularly uranium extraction and milling, and forestry industries.

Table 3.1. Geographic location and surface area of study lakes in (a) the Northwest Territories, and (b) northern Alberta and Saskatchewan.

(a)

Lake	Latitude (deg)	Longitude (deg)	Surface area (km^2)
Colville	67º 10' N	126º 00' W	448
Great Bear	65º 50' N	120º 45' W	31,328
Kelly	65º 23' N	126º 15' W	121
Mirror	64º 51' N	120º 45' W	3
Ste. Thérèse	64º 38' N	121º 35' W	113
Stark	62º 28' N	110º 20' W	274
Willow	62º 10' N	119º 08' W	160
Cli	61º 59' N	123º 18' W	44
Little Doctor	61º 53' N	123º 16' W	21
Trout	60º 35' N	121º 19' W	508

(b)

Lake	Latitude (deg)	Longitude (deg)	Surface area (km^2)
Athabasca	59º 22' N	108º 00' W	7,900
Wollaston	58º 15' N	103º 15' W	2,062
Namur	57º 25' N	112º 40' W	42
Reindeer	57º 15' N	102º 15' W	5,569
la Ronge	55º 30' N	105º 00' W	1,178
Grist	55° 22' N	110° 28' W	25
Cold	54º 33' N	110º 05' W	373
Kingsmere	54º 06' N	106º 27' W	47

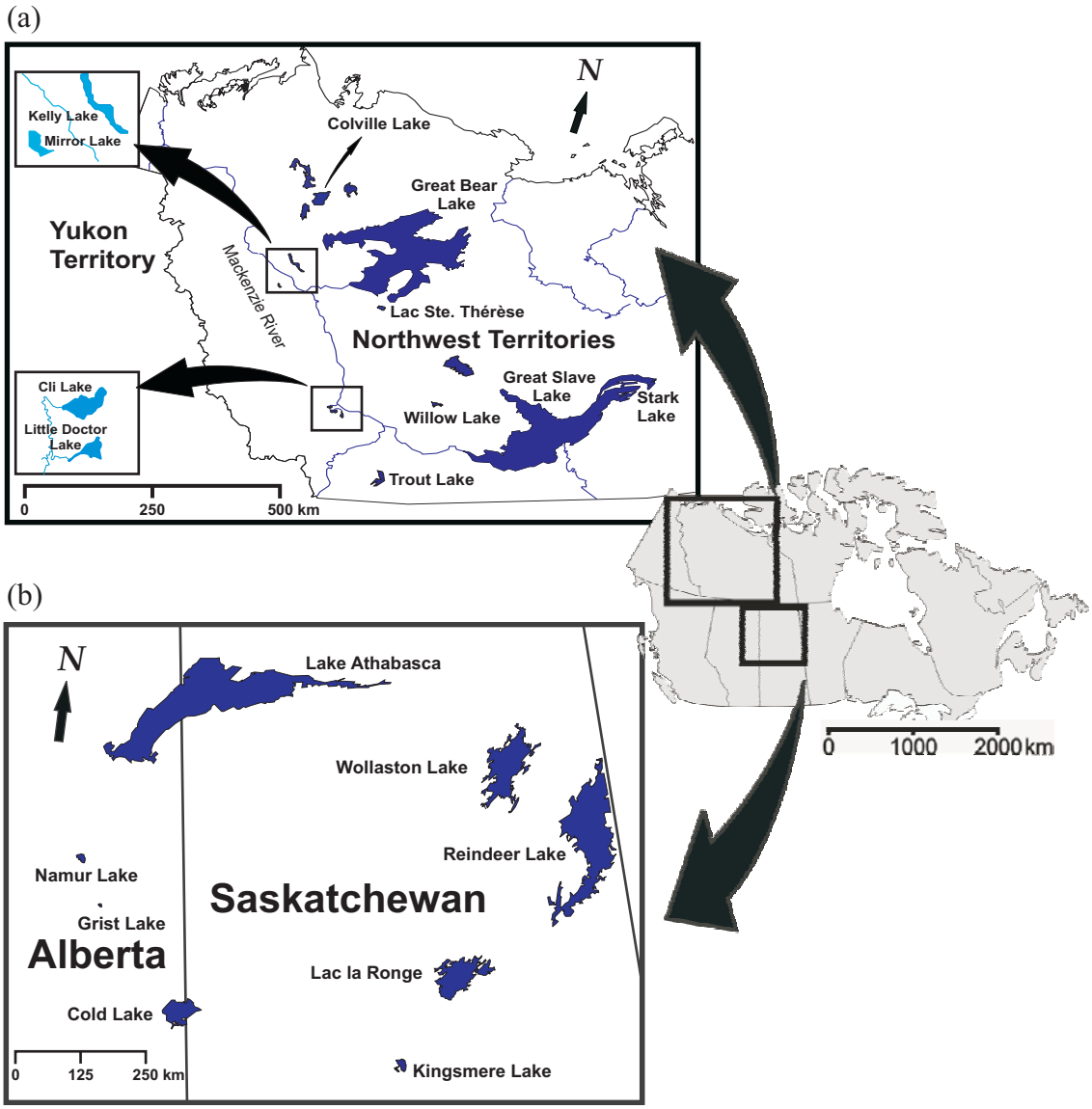


Figure 3.1. Geographic location of the two series of study lakes in (a) the Northwest Territories, and (b) northern Alberta and Saskatchewan, (note difference in scales on the two map insets).

Historically Lake Athabasca was affected by a number of uranium mines, including Beaverlodge mine, which opened in the 1940s. Wollaston Lake and Lac la Ronge are both located in close proximity to both the mining and ore extraction industries, in addition to the forestry industry (Parsons and Barsi 2001). Mining and ore extraction are both sources of mercury to the surrounding environment due waste rock and ore. The surrounding host rock associated with base metal mining is typically enriched in mercury relative to natural background concentrations (Sexauer Gustin *et al.* 2003). Forestry practices have been found to increase both THg and MeHg output from boreal forest catchments (Porvari *et al.* 2003).

The NAS lakes are all mesotrophic drainage lakes with low to moderate TP and algal counts. Additionally, these lakes are characterised by neutral pH and high DO levels throughout the water column (Mitchell and Prepas 1990, Rawson 1936; Muir *et al.* 2001). The Alberta lakes are located on glacial tills while the Saskatchewan lakes, except for Kingsmere Lake, are on the Precambrian Shield. Climate in the NAS region is somewhat warmer compared to the NT region with lake ice cover present six to seven months per year. Lake surface area varies from 25 km^2 (Grist Lake) to 7,700 km^2 (Lake Athabasca) (Figure 3.1(b), Table 3.1(b)). Thus, the NAS lakes are slightly larger than NT lakes, although the median lake surface area in both areas is similar.

3.3.2. Sampling methods

3.3.2.1. Biological sampling

Lake trout collection methods can be found in Section 2.3.2.1. Forage fish, aquatic invertebrates and zooplankton collections were made during the summers of 2001 and 2002 in Cli, Little Doctor, Willow, Mirror, Ste. Thérèse, Kelly and Colville lakes. Stark and Trout lakes were sampled during the summer of 2003. Forage fish species were caught in near shore areas using a beach seine net, or by electro-fishing. Fish were identified and stored frozen in Whirl-Pak® bags. Aquatic invertebrates were collected in littoral areas using common household sieves with a mesh size of 1 to 2 mm. Samples were sorted in the field, frozen in Whirl-Pak® bags and shipped back to National Water Research Institute (NWRI), Saskatoon, where they were later identified by Class and freeze-dried for further analyses.

Zooplankton samples were collected as outlined in section 2.3.2.2.

All data from the NAS lake series was collected as part of a Toxic Substance Research Initiative (TSRI) funded study conducted by Muir *et al.* (2001). Fish from the NAS lakes were obtained from a variety of different sources over 2000 to 2001. Fish from la Ronge, Wollaston, Athabasca, and Reindeer lakes were obtained from the commercial fisheries in the winter of 2000. Grist, Namur and Cold lakes were sampled by Alberta Environment using commercial gill nets of 140 mm in March and October of 2000. Kingsmere Lake was sampled in April of 2000 using commercial gill nets (140 mm) catching 13 lake trout, and in May of 2000 the total sample number was brought to 20 by angling. Whole, frozen fish were shipped whole to NWRI, Saskatoon, for processing.

Lake trout for this study were caught by a variety of methods including gill netting as part of stock assessments and commercial fisheries, and angling. Though pooling of fish caught by different methodology (i.e., gillnetting and angling) has been done in other studies (Muir *et al.* 2001), it can introduce bias into the results. Angling is an active capture method that requires the fish to be biting, and may select for certain age / size ranges depending on distribution within the lake. Also, anglers may want to return with fish of a certain size, not understanding that a large variation is required for a scientific study. Commercial fisheries may forward 'undesirable' fish (i.e., those too large or too small for profitable sale) onto the scientific community, while retaining the 'best of the catch' for sale. Also, gillnetting was done at various times of the year, which can affect fish size because fish do most of their growing in the summer months. These differences, while real, are not crucial to this study in which we compare the features of a similar size range of lake trout and adjust for differences in fish length when comparing ages and mercury concentrations.

Food web sampling in the NAS lakes followed the same methods as previously described for the NT lakes. Sampling of all lakes except for Namur and Grist occurred in June and July 2000; Namur and Grist lakes were sampled in June and July 2001.

3.3.2.2. Mercury analysis on biological tissue

See section 2.3.2.3.

3.3.2.3. Data conversions

Total mercury data for zooplankton and benthic invertebrates were reported in dry weight (dw), while fish were reported as wet weight (ww) concentrations from the analytical laboratories. To convert dw concentrations into ww concentrations, the following equation was used:

$$ww = dw / [(100 - \text{percent moisture}) / 100] \quad (3.1)$$

The percent moisture values were determined through weighing of samples before and after freeze-drying. The data for zooplankton was: mean 90%, range from 86 – 95%, for aquatic invertebrates: mean 83%, range from 77 – 86%.

Methylmercury analyses was performed on NAS zooplankton and aquatic invertebrate samples, in lieu of THg analyses. Therefore, THg estimates in zooplankton were calculated using the proportions of THg to MeHg presented in Plourde *et al.* (1997), i.e., 33%. Total mercury concentrations in aquatic invertebrates were estimated using proportions found by Tremblay and Lucotte (1997) i.e., Odonata 80 %; Gastropoda and Amphipoda 35%.

3.3.2.4. Stable isotope analysis

See section 2.3.2.4.

3.3.3. Statistics

Data normality and variance homogeneity were assessed prior to statistical analysis. Mercury data were log transformed to reduce variance. Linear regression was used to examine the relationship between lake trout age and fork length, and between log THg and $\delta^{15}N$ in the food web. Analysis of variance (ANOVA) was used to detect significant differences between lake trout populations in the two study regions. Homogeneity of slope for the two regions was tested by regression of the two slopes. Maximal Type I error for all analyses was set at $\alpha = 0.05$. Statistics were performed using Minitab 13 statistical software (Minitab Inc, 1999).

3.4. Results

3.4.1. Lake trout population characteristics

Descriptive statistics for lake trout captured in all study lakes are shown in Table 3.2, and raw data for all lake trout sampled are shown in Appendix A. Fork length varied from 470 to 740 mm in the NT study lakes, and from 495 to 740 mm in the NAS lakes. Analysis of variance showed no significant difference in the mean

fork length in these two groups ($p = 0.83$). Lake trout weight ranged from 1,250 to 4,560 g in the NT lakes and 1600 to 4,700 g in NAS lakes. The difference between the mean weights of lake trout from the two areas was not statistically significant ($p = 0.62$). In contrast, the mean age of lake trout in the NT lakes (17.7 years, range 11 to 25 years) was significantly greater than the mean age of lake trout in the NAS lakes (6.7 years, range 5 to 10 years) ($p < 0.001$). Therefore, while lake trout from the two groups were very similar in both fork length and total weight, they differed in age, indicating differences in growth rate.

In the NT lake series, the $\delta^{15}N$ values in lake trout ranged from 9.7 to 16.2 ‰, however, the majority of the values were between 12 and 14 ‰, and a mean value of 12.7 ± 1.6 ‰ (mean ± standard deviation). Values of $\delta^{15}N$ in lake trout from the NAS lakes also ranged from 12 to 14 ‰, with a mean value of 12.9 ± 0.9 ‰. The difference between the mean $\delta^{15}N$ values in the two study areas was not statistically significant ($p = 0.70$).

In the NT lake series, the $\delta^{13}C$ values in lake trout ranged from -25.6 to -30.4 ‰, and in the NAS lakes the values ranged from -23.1 to -28.4 ‰. The difference between $\delta^{13}C$ values for lake trout in the two areas was statistically significant ($p = 0.02$).

The mean (and ± standard deviation) of THg tissue concentration in lake trout from NT lakes was 0.50 ± 0.22 µg Hg/g and the individual lake concentrations ranged from 0.21 to 0.92 µg Hg/g. In comparison, THg concentrations in lake trout from the NAS lakes had a mean (and ± standard deviation) of 0.26 ± 0.07 µg Hg/g, with individual lake concentrations that ranged from 0.14 to 0.41 µg Hg/g.

Total mercury concentration in NT lake trout was significantly greater than NAS in lake trout ($p = 0.02$). In the NT lakes, mercury concentrations in lake trout were significantly correlated with fork length in 50% of the lakes, with weight in 30%, with age in 20%, with $\delta^{15}N$ in 40%, and with $\delta^{13}C$ in 20% of the lakes (Table 3.3a). In the NAS lakes, mercury concentrations in lake trout were significantly correlated with fork length in 87.5% of the lakes, with weight in 87.5%, with age in 37.5%, with $\delta^{15}N$ in 62.5%, and with $\delta^{13}C$ in 12.5% of the lakes (Table 3.3b).

3.4.2. Food webs

The aquatic invertebrate and forage fish species caught in both the NT and NAS lakes are indicated in Table 3.4 and Table 3.5. Mean $\delta^{15}N$ and $\delta^{13}C$ values, as

well as THg concentrations for pooled zooplankton, aquatic invertebrates, forage fish, lake whitefish, and lake trout samples are presented in Table 3.6.

Overall, the stable isotope data showed that food webs from the NT and the NAS lakes were very similar, and lake trout from the two areas have similar diets. However, carbon signatures did differ slightly; Figure 3.2 shows a food web plot of $\delta^{15}N$ and $\delta^{13}C$. Within the NT lakes there was little difference between $\delta^{15}N$ values for zooplankton and aquatic invertebrates. There was a 4 ‰ difference between aquatic invertebrates and the forage fish that are likely feeding on them, and there was approximately a 4 ‰ difference between zooplankton and forage fish, and between forage fish and lake whitefish. There was a 3.1 ‰ difference between lake whitefish and lake trout. Therefore, data show that lake trout are feeding primarily on forage fish rather than invertebrates or feeding as cannibals. Lake trout from the NAS lakes show a similar relationship. Lake trout at the top of the food web pyramid rely on pelagic and periphyton/littoral sources for carbon. Forage fish are feeding in the littoral zones, and their carbon sources are less negative.

Within the NAS lake series, the $\delta^{15}N$ values were nearly 3 ‰ less negative than those sampled from the NT lakes. The difference between forage fish and aquatic invertebrates in the NAS lakes was therefore higher than expected at 4.9 ‰. The difference between zooplankton and forage fish was 3.4 ‰, and the difference between lake whitefish and lake trout was 3.5 ‰.

Differences in $\delta^{15}N$ and $\delta^{13}C$ values between zooplankton, aquatic invertebrates, forage fish, lake whitefish, and lake trout were not statistically significant ($p > 0.05$) for the two regions studied, though trends were evident. Organisms in the NAS lakes were consistently more enriched in $\delta^{13}C$ than organisms from the NT lakes. Nitrogen stable isotope values were similar across the study lakes in both areas, however, in the NAS $\delta^{15}N$ values were higher in the more pelagic species including lake trout, lake whitefish and zooplankton, than in the more near shore species including aquatic invertebrates and forage fish. However, differences in $\delta^{15}N$ values between the two study areas were not statistically different at any level of the food web, as defined by each species.

Table 3.2. Mean (± standard deviation) fork length, weight, age, carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope ratios, and total tissue mercury (THg) concentration in lake trout (*Salvelinus namaycush*) captured in study lakes in the (a) Northwest Territories and (b) northern Alberta and Saskatchewan. Columns give mean ± 1 standard deviation of the stated variable.

Lake	N	Fork Length (mm)	Weight (g)	Age (years)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	THg (µg/g ww)
(a)							
Colville	20	576.4 ± 36.8	1955.0 ± 455.4	11.3 ± 3.1	-25.6 ± 1.3	16.2 ± 0.4	0.21 ± 0.07
Great Bear	10	736.9 ±122.5	4086.4 ±1587.7	19.9 ± 5.1	-27.6 ± 1.5	13.3 ± 1.0	0.35 ± 0.31
Kelly	20	592.8 ± 82.9	2522.0 ± 1177.8	12.3 ± 4.5	-30.4 ± 1.0	12.5 ± 0.4	0.47 ± 0.19
Mirror	20	468.9 ± 41.8	1239.0 ± 287.9	-	-29.8 ± 1.5	9.7 ± 0.6	0.68 ± 0.29
Ste. Thérèse	11	708.1 ± 60.7	3217.9 ± 888.0	24.9 ± 5.2	-28.2 ± 0.7	13.5 ± 0.8	0.77 ± 0.47
Stark	24	726.1 ± 74.4	4559.2 ± 1989.8	23.2 ± 4.6	-26.3 ± 2.4	12.6 ± 0.7	0.47 ± 0.40
Willow	32	613.7 ± 92.9	3067.5 ± 2159.9	15.8 ± 4.8	-27.7 ± 1.1	13.1 ± 0.6	0.38 ± 0.08
Cli	25	499.6 ± 140.1	1651.2 ± 1792.0	16.9 ± 7.2	-26.5 ± 1.2	11.7 ± 1.3	0.92 ± 1.08
Little Doctor	10	547.3 ± 42.3	1603.0 ± 350.0	-	-29.9 ± 0.9	12.3 ± 0.5	0.39 ± 0.08
Trout	28	657.8 ± 67.8	3118.6 ± 1533.1	17.2 ± 5.5	-27.5 ± 1.1	12.3 ± 0.7	0.35 ± 0.08
Mean	**20**	**612.8 ± 93.6**	**2702.0 ± 1104.6**	**17.7 ± 4.8**	**-28.0 ± 1.6**	**12.7 ± 1.6**	**0.50 ± 0.22**
(b)							
Athabasca	20	742.2 ± 57.9	4623.1 ± 1416.9	7.8 ± 1.1	-27.2 ± 1.6	12.0 ± 0.6	0.27 ± 0.07
Wollaston	20	497.5 ± 47.3	1622.7 ± 406.3	5.0 ± 0.6	-26.7 ± 0.8	12.8 ± 0.4	0.14 ± 0.04
Namur	18	580.0 ± 73.2	2396.2 ± 982.5	6.1 ± 1.3	-23.1 ± 0.6	13.4 ± 0.3	0.27 ± 0.14
Reindeer	20	532.1 ± 45.5	1817.0 ± 533.6	5.6 ± 0.8	-27.3 ± 0.6	11.8 ± 0.3	0.41 ± 0.14
la Ronge	19	590.6 ± 185.1	3250.8 ± 2732.8	6.3 ± 1.9	-26.4 ± 0.9	13.9 ± 1.0	0.24 ± 0.16
Grist	18	569.6 ± 95.7	2614.7 ± 1508.9	9.7 ± 3.6	-28.4 ± 0.9	12.1 ± 0.4	0.25 ± 0.20
Cold	16	679.2 ± 70.4	4714.4 ± 1169.1	7.3 ± 1.0	-26.3 ± 0.8	12.9 ± 0.4	0.23 ± 0.08
Kingsmere	20	605.3 ± 66.7	2531.9 ± 750.0	6.0 ± 0.8	-24.8 ± 0.7	14.1 ± 0.5	0.24 ± 0.08
Mean	**19**	**599.6 ± 78.4**	**2946.4 ± 1173.6**	**6.7 ± 1.5**	**-26.3 ± 1.6**	**12.9 ± 0.9**	**0.26 ± 0.07**

Table 3.3. Pearson product moment correlation coefficients (r) for the correlations between log mercury concentrations in lake trout, (*Salvelinus namaycush*), and lake trout fork length, weight, age, and stable isotope ratios ($\delta^{15}N$ and $\delta^{13}C$) in a series of lakes in (a) the Northwest Territories and (b) northern Alberta and Saskatchewan.

(a)

Lake	Fork Length (mm)	Weight (g)	Age (years)	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)
Colville	0.40	0.14	0.35	0.22	-0.41
Great Bear	0.74*	0.62	0.86**	0.89**	-0.42
Kelly	0.47*	0.67**	0.45	0.10	-0.22
Mirror	0.00	0.10	-	0.14	-0.36
Ste. Thérèse	0.24	0.45	0.52	0.92**	-0.30
Stark	0.42*	0.32	0.39	0.51**	-0.81*
Willow	0.00	0.00	0.14	0.35	-0.00
Cli	0.88**	0.83**	0.88**	0.62**	-0.78**
Little Doctor	0.77**	0.69*	-	0.10	-0.26
Trout	0.00	0.10	0.00	0.10	-0.14

(b)

Lake	Fork Length (mm)	Weight (g)	Age (years)	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)
Athabasca	0.20	0.17	0.28	0.62*	-0.10
Wollaston	0.67**	0.59**	0.37	0.10	-0.22
Namur	0.79**	0.74**	0.41	0.35	-0.32
Reindeer	0.49*	0.53*	0.00	0.50*	-0.61**
la Ronge	0.80**	0.85**	0.86**	0.74**	-0.37
Grist	0.83**	0.83**	0.71**	0.75**	-0.14
Cold	0.80**	0.72**	0.53*	0.00	-0.35
Kingsmere	0.82**	0.79**	0.26	0.57**	-0.10

*$p < 0.05$
**$p < 0.01$

Table 3.4. Aquatic invertebrate species caught from lakes in the Northwest Territories (NT) and northern Alberta and Saskatchewan (NAS) during mercury biomagnification studies.

Lake Series	Lake	Date of Capture	Order	Common name
NT	Colville	August 2002	Amphipoda (Gammarus)	amphipod
			Ephemeroptera	mayfly
			Gastropoda	snail
			Hemiptera	boatmen
NT	Great Bear	August 2002	Amphipoda (Gammarus)	amphipod
			Coleoptera	qquatic beetle
			Diptera	fly larvae
			Gastropoda	snail
NT	Kelly	August 2002	Amphipoda (Hyalella)	amphipod
			Gastropoda	snail
			Hemiptera	boatmen
			Trichoptera	caddisfly
NT	Mirror	August 2002	Amphipoda	amphipod
			Ephemeroptera	mayfly
			Gastropoda	snail
			Hemiptera	boatmen
			Trichoptera	caddisfly
NT	Ste. Thérèse	August 2002	Amphipoda (Gammarus)	amphipod
			Hemiptera	boatmen
NT	Stark	September 2003	Corixidae	backswimmer
NT	Willow	August 2000	Amphipoda	amphipod
			Coleoptera	aquatic beetle
			Corixidae	backswimmer
			Gastropoda	snail
			Hemiptera	boatmen
NT	Cli	Summer 1999	Amphipoda	amphipod
			Ephemeroptera	mayfly
			Gastropoda	snail
			Hemiptera	boatmen
			Odonata	dragonfly
			Trichoptera	caddisfly
NT	Little Doctor	August 2000	Amphipoda	amphipod
			Ephemeroptera	mayfly
			Coleoptera	aquatic beetle
			Gastropoda	snail
			Hemiptera	boatmen
			Odonata	dragonfly
			Trichoptera	caddisfly
NT	Trout	September 2003	Amphipoda	amphipod
			Ephemeroptera	mayfly
NAS	Athabasca	July 2000	Bivalva	mussel
NAS	Wollaston	June 2000	Odonata	dragonfly
NAS	Namur	October 2000	Amphipoda (Gammarus)	amphipod
			Amphipoda (Hyalella)	amphipod
			Gastropoda (Fossaria)	snail
NAS	Reindeer	July 2000	Odonata	dragonfly
NAS	Grist	June 2001	Amphipoda (Gammarus)	amphipod
			Gastropoda (Helisoma)	snail
NAS	Cold	September 2000	Gastropoda	snail
NAS	Kingsmere	June 2000	Gastropoda	snail

Table 3.5. Forage fish species caught from lakes in the Northwest Territories (NT) and northern Alberta and Saskatchewan (NAS) during mercury biomagnification studies.

Lake Series	Lake	Date of Capture	Latin name	Common name
NT	Colville	August 2002	*Catostomus catostomus*	longnose sucker
			Coregonus clupeaformis	lake whitefish
			Stenodus leucichthys	inconnu
NT	Great Bear	August 2002	*Cottus cognatus*	slimy sculpin
			Pungitius pungitius	ninespine stickleback
			Salvelinus namaycush	lake trout
NT	Kelly	August 2003	*Cottus cognatus*	slimy sculpin
			Lota lota	burbot
NT	Ste. Thérèse	August 2002	*Esox lucius*	northern pike
NT	Stark	September 2003	*Lota lota*	burbot
NT	Willow	NC	NC	NC
NT	Cli	Summer 1999	*Cottus ricei*	spoonhead sculpin
			Notropis atherinoides	emerald shiner
			Pungitius pungitius	ninespine stickleback
NT	Little Doctor	NC	NC	NC
NT	Trout	September 2003	*Catostomus catostomus*	longnose sucker
NAS	Athabasca	July 2000	*Catostomus cammersoni*	white sucker
			Catostomus catostomus	longnose sucker
			Notropis atherinoides	emerald shiner
			Notropis hudsonius	spottail shiner
NAS	Wollaston	June 2000	*Catostomus catostomus*	longnose sucker
NAS	Namur	October 2000	*Catostomus cammersoni*	white sucker
NAS	Reindeer	July 2000	*Notropis hudsonius*	spottail shiner
			Perca flavescens	yellow perch
NAS	La Ronge	June 2000	*Catostomus cammersoni*	white sucker
			Perca flavescens	yellow perch
NAS	Grist	June 2001	*Notropis hudsonius*	spottail shiner
			Pungitius pungitius	ninespine stickleback
NAS	Cold	September 2000	*Catostomus catostomus*	longnose sucker
			Notropis hudsonius	spottail shiner
NAS	Kingsmere	June 2000	*Notropis hudsonius*	spottail shiner

NC – no forage fish were caught at time of sampling

Table 3.6. Mean (± 1 standard deviation) nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotope values, and total mercury (THg) values for different components of the food web in: (a) Northwest Territory and (b) northern Alberta and Saskatchewan lakes.

Lake Series	Food web level	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)	THg (µg/g ww)
(a)				
NT	zooplankton	3.97 ± 1.30	-32.51 ± 2.30	0.010 ± 0.005
NT	benthic invertebrates	3.77 ± 1.65	-27.58 ± 4.68	0.014 ± 0.010
NT	forage fish	8.46 ± 2.18	-24.19 ± 3.76	0.028 ± 0.017
NT	lake whitefish	9.01 ± 1.36	-25.87 ± 2.99	0.113 ± 0.092
NT	lake trout	12.10 ± 1.59	-27.47 ± 2.48	0.442 ± 0.406
(b)				
NAS	zooplankton	4.46 ± 1.56	-30.09 ± 1.78	0.004* ± 0.002
NAS	benthic invertebrates	2.95 ± 1.16	-24.81 ± 2.91	0.015* ± 0.009
NAS	forage fish	7.87 ± 1.19	-23.44 ± 4.28	0.020 ± 0.010
NAS	lake whitefish	9.36 ± 1.07	-25.67 ± 2.62	0.038 ± 0.031
NAS	lake trout	12.92 ± 0.99	-26.22 ± 1.76	0.257 ± 0.140

* denotes an estimated concentration derived from methylmercury data, as described in section 3.3.2.3.

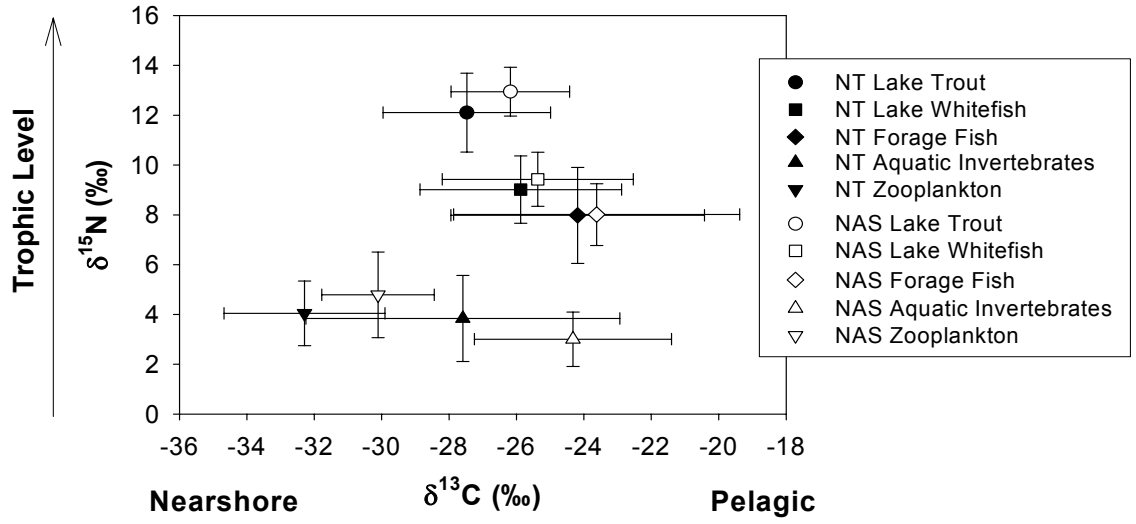


Figure 3.2. Food web plot using data from nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotope analysis. Data points represent mean values calculated from all samples of all aquatic biota species collected in all Northwest Territory and northern Alberta and Saskatchewan study lakes. Error bars represent ± 1 standard deviation.

3.4.3. Mercury biomagnification

Total tissue mercury concentrations increased with increasing $\delta^{15}N$ values in both NT and NAS lakes (Figure 3.3 a and b). This was a significant relationship in all study lakes ($p < 0.01$) although the relationships varied between lakes. Biomagnification rates (defined as the slope of the regression) varied more between lakes in the NT than between those in NAS. In general, slopes were gentler for lakes in the NT than for those in NAS, indicating lower rates of biomagnification in food webs in NT lakes. The intercepts for the regression lines also were higher (Figure 3.3 a and b) in NT than NAS lakes, i.e., organisms at lower trophic levels in the food chain had higher concentrations of total tissue mercury than those in NAS lakes. Data were pooled to compare mercury biomagnification slopes for NT and NAS lakes. The pooled biomagnification slope for the NT lakes was significantly more gentle than that for the NAS lakes ($p < 0.001$) while the y-intercept was significantly higher ($p < 0.001$) (Figure 3.4).

3.5. Discussion

3.5.1. Food web

Trophic relationships can be better understood through $\delta^{13}C$ and $\delta^{15}N$ isotope analyses (Power *et al.* 2002). Together, these two analyses describe both the carbon and nitrogen enrichment through the food web. The food web graph (Figure 3.2) showed a traditional pyramid shape for both the NT and NAS study groups. There was a broad base, showing that lower level organisms feed on a wide variety of carbon sources. Between trophic levels there was approximately a 3 ‰ difference in $\delta^{15}N$ values. High levels of the food web have a narrower range of carbon sources in their diet. Lake trout were the highest trophic level sampled and had the highest $\delta^{15}N$ values indicating that they were primarily feeding on other fish species when possible. The $\delta^{13}C$ values indicate that the food sources of the lake trout came from predominantly pelagic sources. While the differences between the mean $\delta^{13}C$ and $\delta^{15}N$ stable isotopes within all aquatic species collected from the two study areas were not statistically significant, some trends were evident (Figure 3.2).

(a)

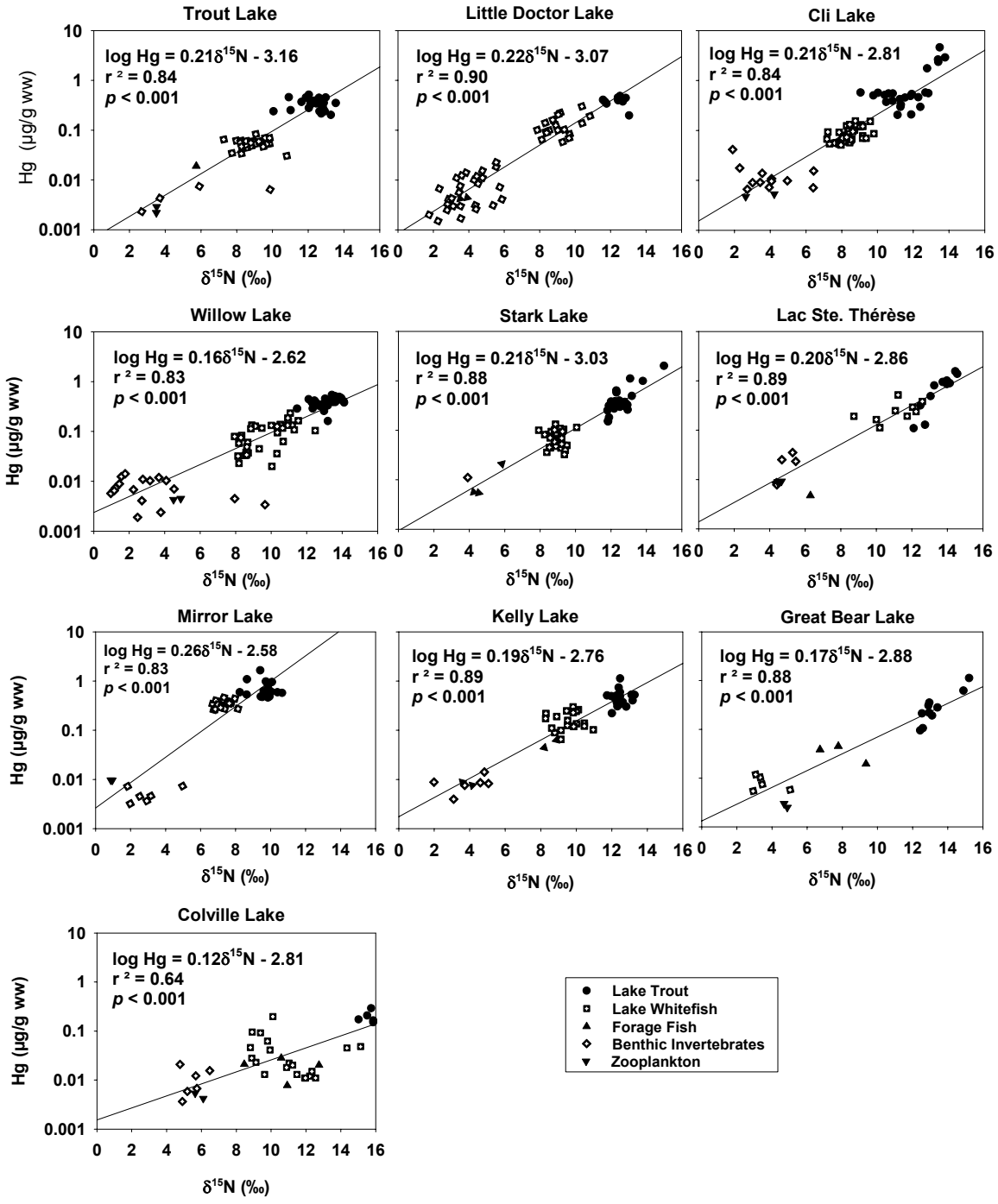


Figure 3.3a. Correlations between nitrogen^{15}isotope values δN^{15} and total mercury in lake trout tissue in lakes in the Northwest Territories. The slope of this relationship is a measure of the rate of biomagnification of mercury through the food chain.

(b)

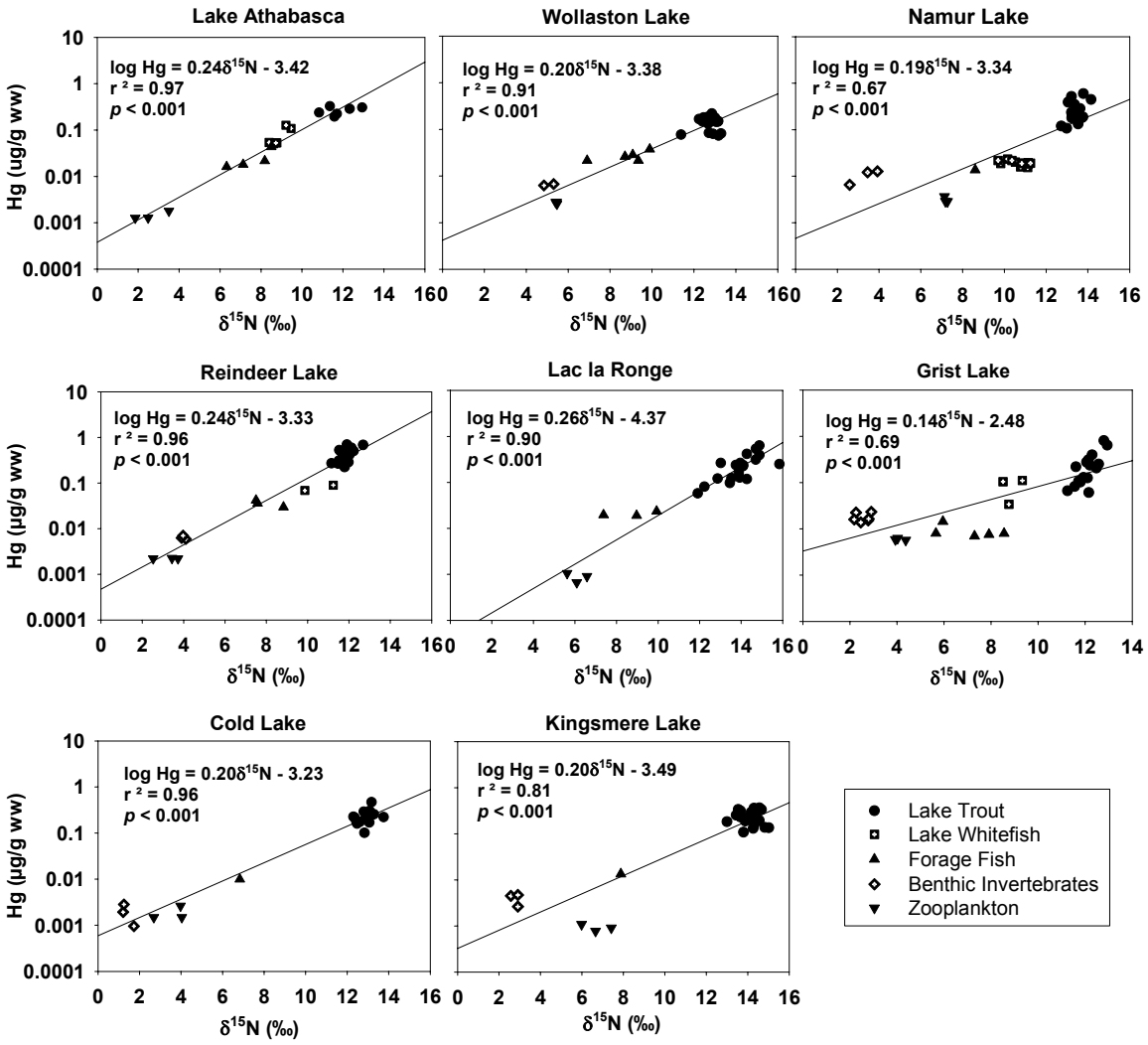


Figure 3.3b. Correlations between nitrogen stable isotope values δN^{15} and total mercury in lake trout tissue in lakes in northern Alberta and Saskatchewan. The slope of this relationship is a measure of the rate of biomagnification of mercury through the food chain.

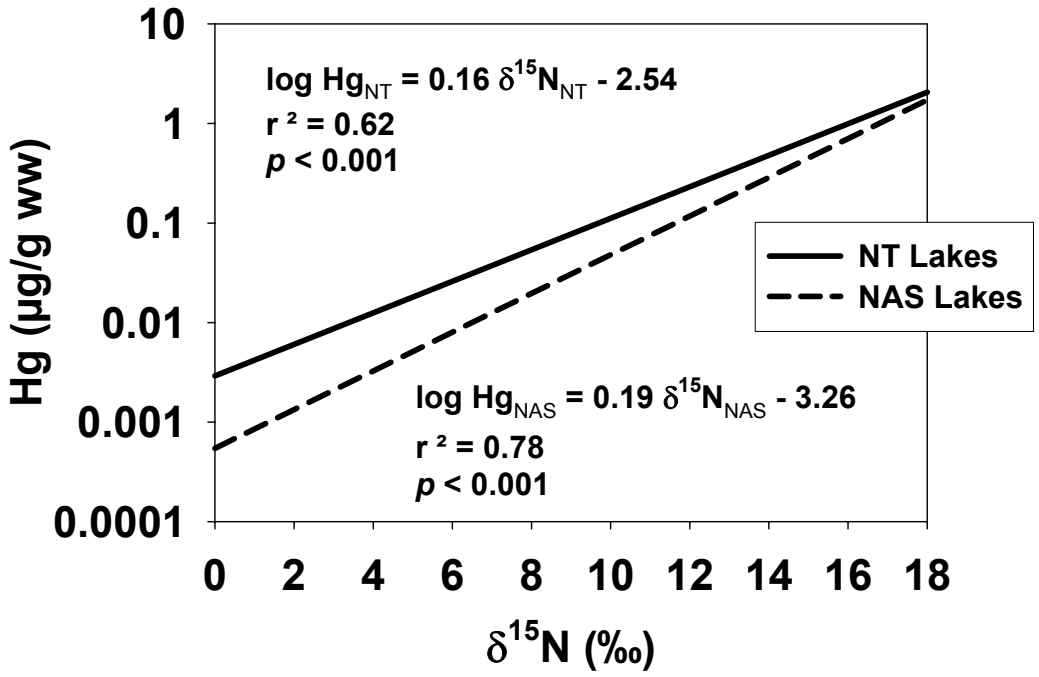


Figure 3.4. Average biomagnification slopes for total mercury in lake trout tissue from pooled data from all lakes in the Northwest Territories (NT) and northern Alberta and Saskatchewan (NAS).

The difference in $\delta^{15}N$ between trophic levels in the NT lakes ranged from 3.1 to 4.0 ‰, and in the NAS lakes from 3.4 to 4.9 ‰. This range of $\delta^{15}N$ values is similar to what has been seen in previous research. Kidd and collaborators (1995) found that in subarctic freshwater lakes the average increase in $\delta^{15}N$ between prey and predator species was approximately 3.3 ‰. Similarly in northern Ontario, Kidd *et al.* (1999) determined a range of 3.8 to 4.3 ‰, and early work done by Peterson and Fry (1987) determined a range of 3 to 5 ‰. The large standard deviation error bars shown associated with the mean isotope in values in Figure 3.2, especially for forage fish and aquatic invertebrates, were likely due to the pooling of various species into one representative sample. Increased sample numbers and analysis by species could reduce this error in future studies.

3.5.2. Mercury biomagnification

The biomagnification rate estimates the relative increase in mercury per unit $\delta^{15}N$ based on contamination at the base of the food web rather than in the water column (Broman *et al.* 1992). The antilog of the biomagnification rate is called the food web magnification factor (FWMF) (Fisk *et al.* 2001).

This study found a significant, positive correlation between trophic level, measured as $\delta^{15}N$, and the log total tissue mercury concentration in all NT and NAS lakes investigated. This positive correlation was expected as many previous studies have shown similar correlations in study areas ranging from the Canadian subarctic to Lake Malawi, East Africa (Cabana and Rasmussen 1994; Kidd *et al.* 1995, 2003; Power *et al.* 2002). The biomagnification rates in the NT lakes ranged from 0.12 to 0.24 µg Hg/g in tissues per 1 ‰ $\delta^{15}N$, while the biomagnification rates in the NAS lakes ranged from 0.16 to 0.33 µg Hg/g in tissues per 1 ‰ $\delta^{15}N$ (Figure 3.3 a. and b.).

The biomagnification rates found in this study are similar with those found in other studies that have been done in the past. Kidd *et al.* (1995) found values ranging from 0.09 to 0.16 µg Hg/g in tissues per 1 ‰ $\delta^{15}N$ in north western Ontario lakes. Also, Power *et al.* (2002) found a biomagnification rate of 0.19 µg Hg/g in tissues per 1 ‰ $\delta^{15}N$ in Stewart Lake, located in the eastern Arctic. Though the location of Stewart Lake is closer to the NT lake latitudes, the fish sampled by Power *et al.* (2002) were

smaller and younger than those found in the NT lakes, which could have resulted in the lower rate of biomagnification.

The food web magnification factor for the NT and NAS lakes ranged from 1.32 to 1.74, and 1.45 to 2.14 respectively. Muir *et al.* (2001) compared contaminant biomagnification in aquatic food webs from lakes across temperate North America based on FWMFs. Muir *et al.* (2001) found that the lowest FWMFs came from Opeongo Lake in New York State (1.49), but there was also high variation among the lakes in this study; nearby Seneca Lake had a higher FWMF of 1.96. Values in northwest Ontario ranged from 1.49 to 1.71 (Muir *et al.* 2001).

The lower biomagnification rate associated with pooled data from the NT lakes than the NAS lakes (Figure 3.4) suggests a slower rate of biomagnification of mercury in aquatic food webs in the more northern lakes. It has already been determined that there was no significant difference in the $\delta^{15}N$ isotope values at each trophic level, including lake trout, between the two areas. Therefore, another explanation must be found for the differing biomagnification rates. The difference may be a reflection of slower growth rates in the north where colder climates can lead to slower metabolism and decreased feeding (Wootton 1990; 1992). The rate of food web biomagnification of many contaminants may be related to growth rates (Muir *et al.* 2001; Schinlder 1995) and to low light, low temperature, and low nutrient concentrations which affect growth rates (Muir *et al.* 2001). Schindler (1995) attempted to explain the variations in biomagnification rates as a reflection of other ecological and physical factors including differential feeding ecology and differences in individual metabolic rates leading to slower growth in colder climates.

Slower growth rates, when combined with the greater mean age of lake trout in the NT lakes, could explain the higher THg concentrations. Age clearly is an important factor in the variation of mercury concentrations in fish, mercury biomagnification rates between the two study areas, and the eventual convergence of the biomagnifications slopes as the age of the fish reaches 20 years (Figure 3.4). Although the lake trout from the NT lakes were older than those in the NAS lakes, the two groups of lake trout were of similar size, and had similar prey items, as shown in Figure 3.2.

A second important factor affecting higher mercury levels in NT than NAS lake trout relates to baseline mercury concentration, as defined by the y-intercepts; these intercepts generally were higher in NT food webs than in NAS food webs (Figures 3.3 a. and b., Figure 3.4). Reasons for higher mercury concentrations in lower trophic levels in the NT lakes remains unknown at this time since there was no difference in mercury concentration in the water column, and both areas have both low THg and low MeHg concentrations in the water (Chapter 2, Muir *et al.* 2001). Some differences may be associated with lake size; NAS lakes tend to be somewhat larger than NT lakes with lower dissolved organic carbon (DOC) concentrations.

In addition to the effect of trophic relationship on mercury biomagnification, carbon sources are important with organic contaminants accumulating in greater concentrations in pelagic food webs than in littoral food webs (Campbell *et al.* 2003; Gorski *et al.* 2003). Tremblay (1999) found that fish feeding on aquatic insects ingest organisms with lower concentrations of THg in their tissues compared to fish feeding on zooplankton from the pelagic food web. No significant difference was found between the mean $\delta^{13}C$ values in any level of the food webs evaluated from the two regions (Figure 3.3). However, there was a trend, although not significant, showing that the organisms collected from the NAS lakes were consistently more enriched in $\delta^{13}C$ than those from the NT lakes (Table 3.2). More negative $\delta^{13}C$ values are associated with more pelagic, food sources, while more positive values are associated with more littoral, food sources (Hecky and Hesslein 1995). Therefore, our findings infer that the food webs in the NT lakes are more pelagic in origin. Past research (Campbell *et al.* 2003; Power *et al.* 2002; Gorski *et al.* 2003) has reported that more negative $\delta^{13}C$ values are correlated with higher mercury concentrations, and while the present study has no statistical proof of such a relationship, the trend was present in the data. This study found that higher mean tissue THg concentrations were found in the NT lake food webs, and that these food webs also had more negative $\delta^{13}C$ values when compared with the NAS lake food webs. Further work, with more detailed sampling at each level of the food web, may provide more statistical certainty to this argument.

3.6. Conclusions

Lake trout feeding as inferred from an investigation of $\delta^{15}N$ relationships in the food web, were similar in NT and NAS lakes, i.e., lake trout from both areas appeared to feed predominantly on insectivorous fish species. The differences found in feeding habits do not explain the differences in THg concentration in the tissue of the lake trout. Log mercury concentrations in biota were positively correlated to $\delta^{15}N$ values in all study lakes. Biomagnification rates were lower in the NT lakes compared to the NAS lakes. Lake trout were substantially older in NT than NAS lakes, suggesting that their higher mercury concentrations were due to greater mercury accumulation, albeit at a lower annual rate, over their longer lifetime. The intercept for the slopes was higher for NT than NAS lakes suggesting greater mercury concentrations at the base of the food web. This could be due to slightly smaller NT than NAS lakes with higher DOC concentrations. Carbon isotope analyses showed that the NT lake food webs were consistently (but not significantly) more pelagic in nature as evidenced by more negative $\delta^{13}C$ values, than NAS lakes which relied more on littoral zone carbon. Therefore, higher mercury burdens found in the NT lake trout might be in part due to the nature of the more pelagic food web. Further detailed study of the food web components with greater sample sizes may improve the statistical significance of these relationships.

4.0 GROWTH RATE AS A FACTOR AFFECTING MERCURY CONTAMINATION IN LAKE TROUT, *SALVELINUS NAMAYCUSH*, IN TWO SERIES OF LAKES IN NORTHERN CANADA

4.1. Abstract

Past research has shown an inverse correlation between growth rate and mercury contamination in a variety of fish species from different geographical areas. Higher total mercury (THg) concentrations in muscle tissue have been correlated with slower growth rates. In this study two different groups of lake trout (*Salvelinus namaycush*), one from a series of lakes in the Northwest Territories (NT), the other from a series of lakes in northern Alberta and Saskatchewan (NAS), were compared with respect to growth rate and total tissue mercury concentration. There was a significant, inverse correlation between growth rate and mercury contamination in lake trout, both in the NAS study area ($p = 0.024$) and in the NT study area ($p < 0.001$). When the two study areas were compared, mercury concentrations were higher in lake trout in the NT lakes, than lake trout from the NAS lakes. Also, lake trout from the NT lakes were slower growing than from the NAS lakes ($p < 0.001$). Possible explanations for the difference in growth rates between the two areas include climate differences, which can affect metabolic rates and lake productivity, and differences in fishing pressure, which can affect population structure and crowding.

4.2. Introduction

Many remote northern lakes in North America support piscivorous fish species with mercury levels exceeding concentrations considered safe for human consumption (0.5 to 1.0 µg Hg/g) (Weiner and Spry 1996; Lockhart *et al.* 2005). Mercury is taken up from the environment by fish at a faster rate than it is eliminated, leading to the continual bioaccumulation of mercury in fish tissues. Many factors affect the bioaccumulation of mercury in lake trout. Mercury concentration in the tissue of various predaceous freshwater fish species often is positively correlated with fish age

and length (Bodaly *et al.* 1993; Grieb *et al.* 1990; Vander Zanden and Rasmussen 1996; Evans *et al.* 2005). Moreover, in the Mackenzie River Basin (MRB) high mercury concentrations in lake trout are strongly and positively correlated with fish age and inversely correlated with lake surface area (Evans *et al.* 2005; Chapter 2). Mercury concentrations in MRB lake trout are also positively correlated with fork length and watershed-to-lake surface area ratio and inversely correlated with latitude (Chapter 2).

Previous research has shown that mercury levels are higher than in lake trout from lakes in the western Northwest Territories area higher than in lake trout from northern Alberta and Saskatchewan (NAS) lakes (Muir *et al.* 2001; Evans *et al.* 2005). Furthermore, biomagnification rates from plankton through to lake trout are slower for NT compared to NAS lakes (Chapter 3). Lake trout were of the same approximate size from both locations; however, they were older in NT than NAS lakes. Previous studies have suggested that biomagnification is affected by growth rate (Muir *et al.* 2001; Schindler *et al.* 1995). It is of interest, therefore, to determine the extent to which lake trout growth rates from the two study areas are different and how this could affect mercury bioaccumulation.

Growth rates of lake trout, as in other species, can vary with respect to geography. Lake trout in more northern regions are often slower growing than those from lakes in central Canada (Scott and Crossman 2001). Lake trout from Lac la Ronge, Saskatchewan (SK), had a mean fork length of 660 mm at age ten, while fish of the same age from Great Bear Lake, NT, had a mean fork length of 372 mm (Scott and Crossman 2001). Lake trout from more northern regions reach the age of sexual maturity slower than other populations, and this is likely related to size (Scott and Crossman 2001). Lake trout generally reach sexual maturity at age six to seven, while sexual maturity at age 13 is not uncommon in Great Bear Lake (Scott and Crossman 2001). Therefore, lake trout living in the Northwest Territories are generally are older and slower growing than lake trout from more southern locations.

Growth rates are affected by a number of variables, with low water temperature likely a major cause of the slower growth rates Observed in more northern lakes. Water temperature affects both food consumption and metabolism (Wootton 1990; 1992). Fish living in sub-polar or temperate regions, including the lake trout studied here, grow

slowly or not at all during winter months, and then very quickly through the spring and summer months (Wootton 1990). Lakes in the NT are ice covered for longer than those in more southern areas, leading to a shorter growing season.

Fish with slower growth rates have higher metabolic turnover per unit of increase in body weight (Verta 1990). Stafford and Haines (2001) proposed biodilution, decreasing mass-specific concentrations with increased biomass (Thomann 1989), of mercury throughout the tissues as a possible explanation. This explains why fish with a faster growth rate often have lower mercury concentrations than slower growing fish (Olsson 1976; Huckabee *et al.* 1979). Nevertheless, old, slow growing fish may have high mercury levels because of their longer lifespan over which they bioaccumulate mercury; growth dilution of methylmercury (MeHg) is not sufficiently rapid to offset this effect (Rose *et al.* 1999).

Fish harvesting pressures can be important factors affecting growth rates. The abundance of fish in a lake is determined by recruitment, natural mortality and mortality due to fishing pressures. Fish tend to be more abundant in lakes where there is low emigration, and natural mortality is more common than mortality due to fishing pressures. Fishing pressures are typically directed to larger fish; by removing larger fish competition for food is reduced allowing younger fish to be more abundant and faster growing. Fishing pressures tend to be light in the majority of NT lakes where the human population is sparse, making relatively small subsistence demands on these lakes. In contrast, lakes in the NAS are surrounded by a larger human population with intensive sport, and often commercial fisheries.

Fishing pressures can affect mercury levels in fish by affecting growth rates. This was shown in a study by Verta (1990) who measured growth rates before and after an intensive fishing operation in a remote Finnish lake in which half of the lake's fish biomass, including northern pike (*Esox lucius*), burbot (*Lota lota*), and roach (*Rutilus rutilus*), was removed. Northern pike, a predaceous species, growth rates doubled and mercury concentrations fell following the intensive fishing operation. Northern pike also became more reliant on zoobenthos since the population of zoobenthos increased following the period of intensive fishing. This shift in diet could have affected both the growth rate and mercury intake.

The overall objective of this chapter is to continue exploring the reasons why lake trout in NT lakes have higher mercury concentrations than NAS lakes. In Chapter 2, it was shown that mercury biomagnified at a slower rate in NT than NAS lakes although mercury concentrations at the base of the food web (as inferred from the regression slope) were higher in NT than NAS lakes. Some differences appeared to be related to the biology of the lake trout, specifically their age and size. The first objective of this research chapter was to compare lake trout growth rates in a series of NT lakes in the Mackenzie River Basin (MRB) with the growth rates of lake trout in a series of NAS lakes. The second objective of this research chapter was to determine if differences in growth rates can help explain differences in total mercury (THg) concentrations in lake trout tissue between the two areas of study.

4.3. Materials and Methods

4.3.1. Study areas

The NT study area in this chapter mirrors that of Chapter 2. The NAS study area mirrors that described in Chapter 3, with the addition of Wassegam Lake, SK. For background information on the two study areas, refer to Section 3.3.1.

The lakes in the NT study area are located in the MRB, along both sides of the Mackenzie River from Trout Lake in the south to Aubry Lake in the north (Table 4.1a, Figure 4.1a). The climate is sub-arctic with mean annual temperatures averaging from -5.4 to 8.7°C (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/results_e.html, accessed January 18, 2005) (Table 4.2). The MRB has a very low human population density. With the exception of Great Slave Lake, there are no commercial fisheries on any of the NT study lakes. Great Bear Lake supports a very active sport fishing industry, and the other lakes see limited sport fishing and subsistence fishing from 'First Nation' communities in the vicinity.

The NAS lake series is comprised of lakes located in the central and northern areas of Alberta and Saskatchewan (Table 4.1b, Figure 4.1b). Mean annual air temperature throughout the NAS study area ranges from -0.1 to 2.1°C (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/index_e.html, accessed January 18, 2005) (Table 4.2). The climate surrounding the NAS lakes is somewhat warmer than that in the area surrounding the NT lakes. The lakes are ice covered six to

Table 4.1. Geographic location and surface area of study lakes in both (a) Northwest Territories and (b) northern Alberta and Saskatchewan**.**

(a)

Lake	Latitude (degrees, minutes)	Longitude (degrees, minutes)	Surface area (km^2)
Aubry	67° 24’ N	126° 27’ W	380
Colville	67º 10’ N	126º 00’ W	448
Manuel	66° 58’ N	128° 54’ W	52
Rorey	66° 55’ N	128° 24’ W	60
Belot	65° 53’ N	126° 16’ W	304
Great Bear	65º 50’ N	120º 45’ W	31328
Turton	65° 48’ N	128° 24’ W	48
Mahony	65° 30’ N	125° 20’ W	181
Kelly	65º 23’ N	126º 15’ W	121
Ste. Thérèse	64º 38’ N	121º 35’ W	113
Stark	62º 28’ N	110º 20’ W	274
Willow	62º 10’ N	119º 08’ W	160
Great Slave	62° 50’ N	113° 50’ W	28568
Cli	61º 59’ N	123º 18’ W	44
Trout	60º 35’ N	121º 19’ W	508

(b)

Lake	Latitude (degrees, minutes)	Longitude (degrees, minutes)	Surface area (km^2)
Athabasca	59º 22’ N	108º 00’ W	7900
Wollaston	58º 15’ N	103º 15’ W	2062
Namur	57º 25’ N	112º 40’ W	42
Reindeer	57º 15’ N	102º 15’ W	5569
la Ronge	55º 30’ N	105º 00’ W	1178
Grist	55° 22’ N	110° 28’ W	25
Cold	54º 33’ N	110º 05’ W	373
Wassegam	54° 17’ N	106° 14’ W	10
Kingsmere	54º 06’ N	106º 27’ W	47

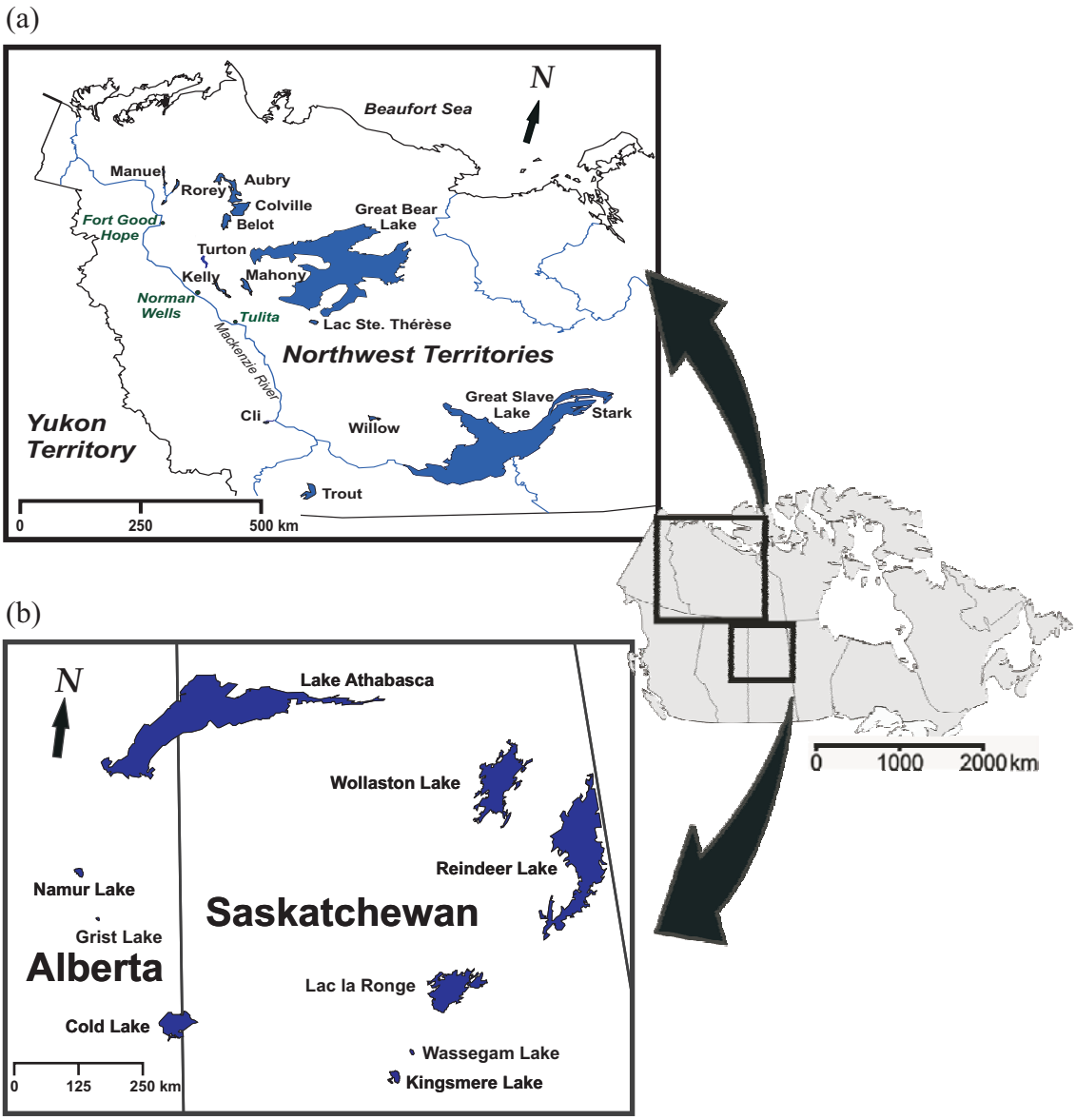


Figure 4.1. Map of study lakes in (a) the Northwest Territories, Canada, and (b) northern Alberta and Saskatchewan, Canada.

Table 4.2. Selected climate variables from Environment Canada weather stations located within the Northwest Territories (NT) and northern Alberta and Saskatchewan (NAS) study areas. Climate variables included average annual temperature, the number of days when maximum recorded temperature was > 0°C, the number of days when minimum temperature was less than or equal to 0°C, and the average number of months when study lakes in that area were ice-covered (http://www.climate.weatheroffice.ec.gc.ca/climate_normals/index_e.html, accessed January 18, 2005).

Weather Station	Average Temperature (°C)	No. days with max. temp. >0°C	No. days with min. temp. </= 0°C	No. months when lakes are ice-covered
NT				
Inuvik	-8.8	106.7	258.5	7 - 8
Norman Wells	-5.5	181	232.6	7 - 8
Yellowknife	-4.6	190	222.2	7 - 8
NAS				
Athabasca	2.1	262.1	200.6	6
la Ronge	-0.1	236.5	213.9	6 – 7
Cold Lake	1.7	254.8	199.2	6

seven months per year (Table 4.2). Lake surface area ranges from at 10 km^2 (Wassegam Lake) to 7,700 km^2 (Lake Athabasca) (Figure 4.1(b), Table 4.1(b)). Many of the lakes in the NAS study area support active fisheries. Lakes Athabasca, Wollaston, Reindeer and la Ronge all have commercial fisheries. Cold, Namur and Grist lakes are all popular with anglers, and support sport fishing industries. Grist Lake, in Alberta, has somewhat limited road access. Kingsmere and Wassegam lakes are located within Prince Albert National Park (PANP) boundaries, and have restrictions on lake trout fishing. Kingsmere Lake is popular with anglers while Wassegam is rarely fished as it is remote and difficult to access.

4.3.2. Sampling methods

4.3.2.1. Biological sampling

With the exception of Wassegam Lake, biological sampling methods have already been discussed in section 3.3.2.1 and are not repeated here. For Wassegam Lake, 20 lake trout were caught by angling along the northwest shore from June 10 to 12, 2003. The fish were weighed, and fork length was measured. Fish were sexed and a sample (approximately 30 g) of dorsal muscle tissue was removed for mercury and stable isotope analysis. These samples were placed in small Whirl Pak® bags, and kept on ice for three days until return to the lab. Sub-samples (20 g) were then sent to the Freshwater Institute (FWI) in Winnipeg for mercury analysis. Both sagittal otoliths were removed in the field and preserved in scale envelopes for age determination.

4.3.2.2. Mercury analysis on biological tissue

Refer to section 2.3.2.3

4.3.3. Statistical methods

Lake trout data from all NT lakes were pooled. Likewise, all lake trout data from the NAS lakes were pooled. Growth rates were calculated by regressing fork length on lake trout age for all lake trout in NT lakes, and for all lake trout in NAS lakes. Differences in mean growth rate between the NT and NAS study areas was investigated through the use of the student's t-test. Maximal Type I error for all analyses was set at $\alpha = 0.05$.

Statistical methods used to determine whether differences in THg tissue concentration were related to differences in growth rate followed those employed by Stafford and Haines (2001). Log age (years) was regressed on fork length (mm) and the negative residuals of this regression were used to obtain a growth rate index. Next, log total mercury (μg Hg/g wet weight (ww)) was regressed on fork length (mm). The residuals of this regression were used to obtain a size-normalized mercury concentration index. The mercury concentration residuals (MCRs) were then regressed versus the growth rate residuals (GRRs) to investigate the relationship between mercury concentration and growth rate in the lake trout after accounting for length. Statistical significance of the relationships was judged using *F*-statistics calculated for the regression, and *p*-values. Maximal Type I error for all analyses was set at $\alpha = 0.05$. All statistics were performed using Minitab 13 statistical software (Minitab, Inc. 1999).

4.4 Results

4.4.1. Comparison of growth rates

The regression equation describing growth rate for lake trout in NT lakes was:

$$\text{length}_{NT} = 10.45 \times age_{NT} + 443.99,\ r^2 = 0.32. \text{ (Figure 4.2)} \quad (4.1)$$

The regression equation describing lake trout growth rate in the NAS lakes was:

$$\text{length}_{NAS} = 35.06 \times age_{NAS} + 360.28,\ r^2 = 0.34 \text{ (Figure 4.2)} \quad (4.2)$$

The slope of the growth regression for the NAS lakes was significantly steeper than the slope for the NT lakes ($p < 0.001$) (Figure 4.2). From the graph, the estimated age of a 600 mm lake trout was 14.9 years in the NT lakes, and 6.8 years in the NAS lakes. Lake trout in NAS lakes grew at an average of 35.1 mm per year, while those in the NT lakes grew at an average of 10.4 mm per year. The steeper slope associated with the NAS lakes indicates that lake trout from these lakes grew faster than their northern counterparts in the NT lakes. There was a larger age and size range in NT than NAS lake trout.

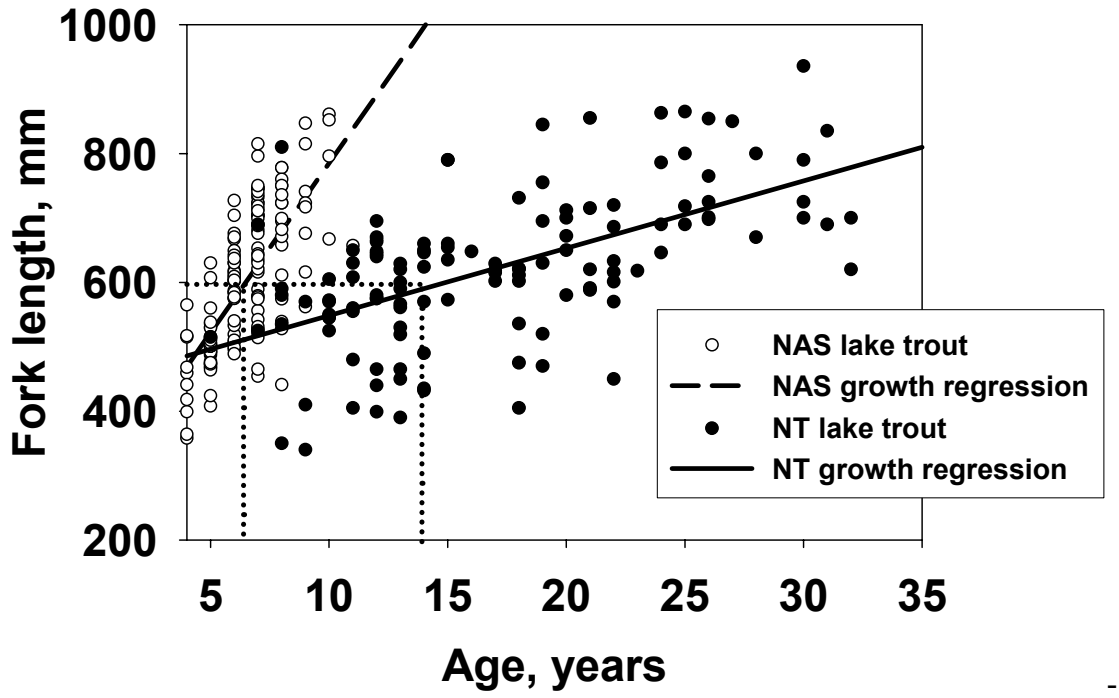


Figure 4.2. Growth regressions for groups of lake trout collected from lakes in the Northwest Territories (NT) and in northern Alberta and Saskatchewan (NAS). The dotted lines represent age at a standardized fork length of 600 mm for both populations. $p < 0.001$ for both regressions.

4.4.2 Correlation between growth rate and total mercury concentration

The relationship between log age and fork length for NT lake trout was

log age = 0.0071 × *fork length* + 0.716 (r^2 = 0.18, $p < 0.001$) (Figure 4.3a). (4.3)

For NAS lake trout, the relationship between age and fork length was

log age = 0.0006 × *fork length* + 0.465 (r^2 = 0.26, $p < 0.001$) (Figure 4.3b). (4.4)

Lake trout from Wassegam and Grist lakes stand out above the regression line, as these fish were old for their fork length when compared to lake trout from the other NAS lakes. Therefore, the growth residuals associated with these fish are larger than those seen in fish from other lakes.

The slope of the log age and fork length relationship was slightly, but not significantly, steeper in the NT lakes (0.0007) than in the NAS lakes (0.0006) (p = 0.438). Therefore, lake trout in NT lakes were growing slower when compared to lake trout from the NAS lakes. The intercepts, however, were significantly different (p = 0.002). At the lower end of the sampled fork length range (approximately 300 mm), the age of lake trout in the NT calculated from the regression line was 10.4 years. This is almost double the age of lake trout from the NAS lakes calculated from the regression line, which was 5.8 years.

The relationship between log mercury concentration and fork length for the NT lakes is described by the equation

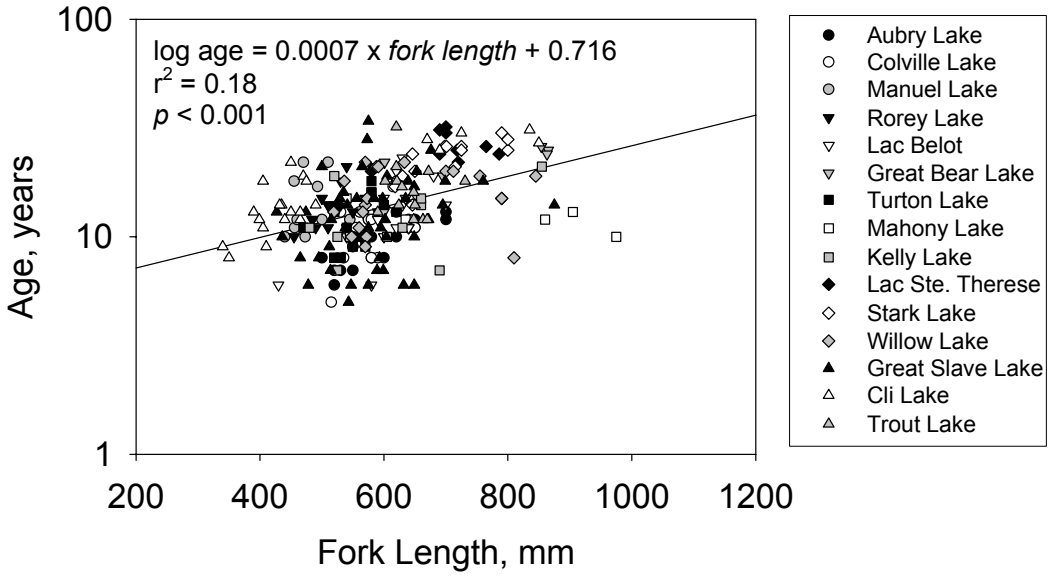
log Hg = 0.0008 × *fork length* – 0.982 (r^2 = 0.10, $p < 0.001$) (Figure 4.4a). (4.5)

The estimated mercury concentration in the muscle tissue of a lake trout with a fork length of 600 mm is 0.31 µg Hg/g ww. Lake trout from Cli Lake (Figure 4.4a) and to a lesser degree Lac Ste. Thérèse stand out with particularly high tissue mercury concentrations with respect to fork length. The relationship between log THg concentration and fork length in lake trout from the NAS lakes is

log Hg = 0.0011 × *fork length* – 1.260 (r^2 = 0.26, $p < 0.001$) (Figure 4.4b). (4.6)

Therefore, the estimated mercury concentration for a 600 mm lake trout in the NAS lakes is 0.25 µg Hg/g ww. Lake trout from Reindeer Lake had the highest mercury concentrations.

(a)



(b)

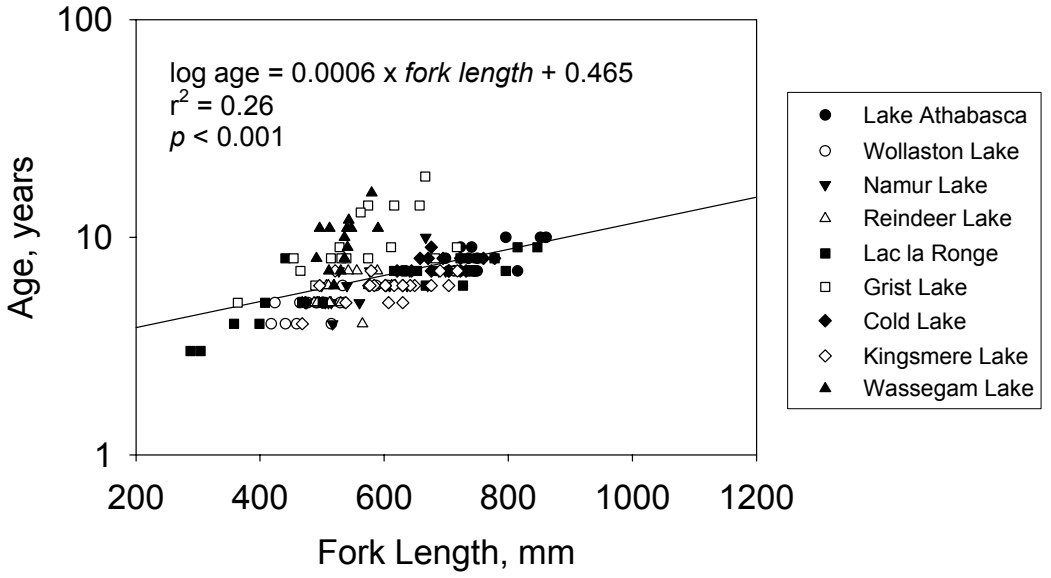
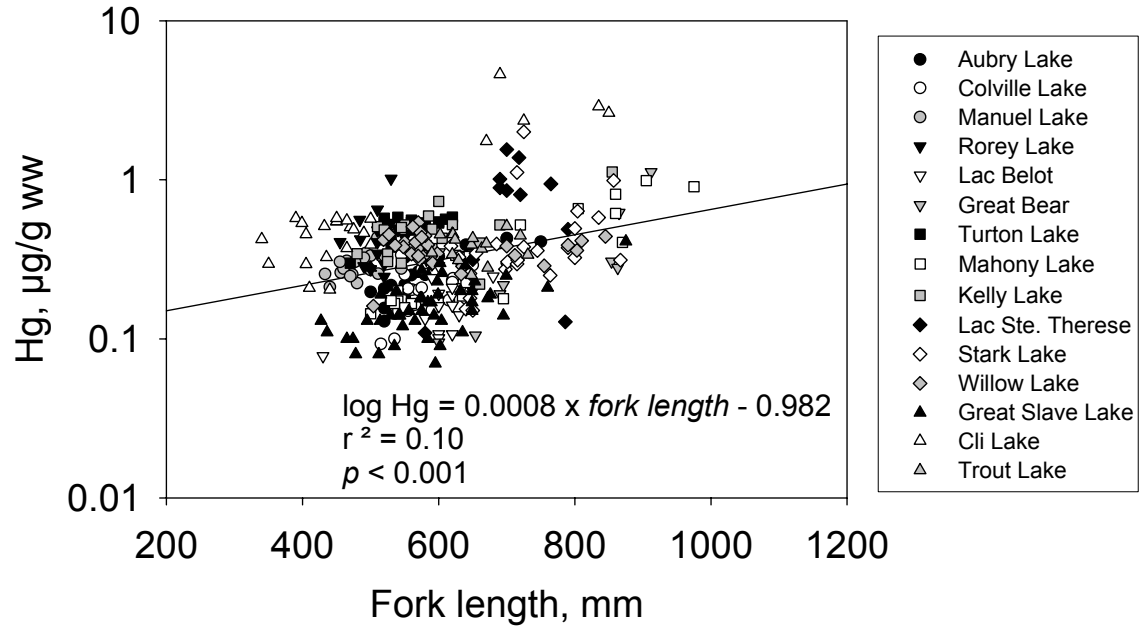


Figure 4.3. Regression of age vs. fork length in lake trout from study lakes in (a) the Northwest Territories and (b) northern Alberta and Saskatchewan.

(a)



(b)

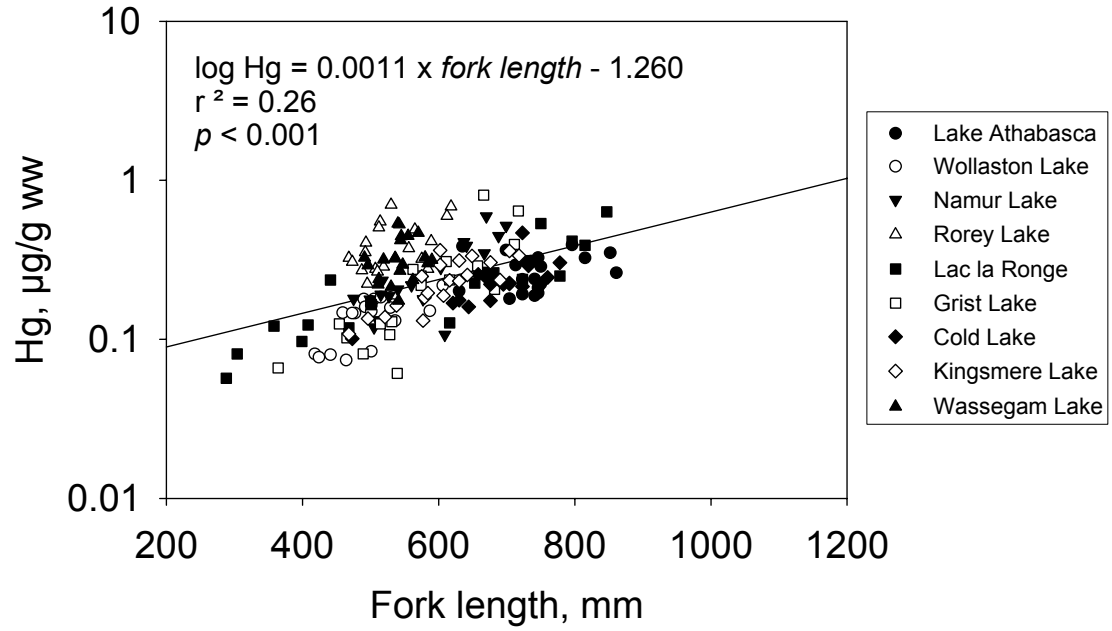


Figure 4.4. Regression of total mercury in lake trout tissue versus fork length of lake trout in study lakes in (a) the Northwest Territories, and (b) northern Alberta and Saskatchewan.

While the relationship between mercury concentration and fork length is significant in both study areas, there is a stronger relationship in the NAS lakes. The intercept of the relationship associated with the NT lakes is significantly higher (0.16 µg Hg/g) than that of the NAS lakes (0.06 µg Hg/g) ($p = 0.028$). The mercury concentrations were highest in the smallest NT fish (approximately 300 mm) and were higher than they were in lake trout of a similar size from the NAS lakes. Also of interest is that even the smallest fish sampled in the NT lakes were nearly double the age (8.4 years) of those of the same length in the NAS lakes (4.4 years).

The residuals of the log mercury versus fork length (MCRs) were regressed on the negative residuals of log age versus fork length (GRRs) for both the NT and NAS lakes (Figure 4.5). There was a statistically significant negative relationship between MCR and GRR described by the equation

$$\text{MCRs} = -0.563 \times GRRs - 0.033 \ (r^2 = 0.11, p < 0.001) \quad (4.7)$$

Cli Lake fish, with their higher than average mercury concentrations and slower then average growth rates appear in the top left quadrant of the plot in Figure 4.4a, high above the regression line.

For NAS study lakes, there was a statistically significant relationship between mercury and growth rate (Fig. 4.5). The equation for that relationship is

$$\text{MCRs} = -0.302 \times GRRs - 0.009 \ (r^2 = 0.03, p = 0.024) \quad (4.8)$$

Lake trout from Reindeer Lake stand out in the upper right hand quadrant above the regression in Figure 4.5b. These fish had high mercury concentrations, and grew at a faster rate when compared to lake trout from other NAS lakes in the region. The dotted regression line in Figure 4.5b shows the same correlation with the lake trout from Reindeer Lake removed. The resulting equation is

$$\text{MCRs}_{NR} = -0.500 \times GRRs_{NR} - 0.054 \ (r^2 = 0.13, p < 0.001). \quad (4.9)$$

There was a statistically significant inverse correlation between the mercury concentration in lake trout muscle tissue and growth rate in both study areas. Although the slope of the relationship between mercury concentration and growth rate appears steeper in the NT lakes (Figure 4.5a) when compared to the NAS lakes (Figure 4.5b), differences are not statistically significant ($p = 0.167$). Also, differences in the y-intercept are not statistically significant ($p = 0.316$).

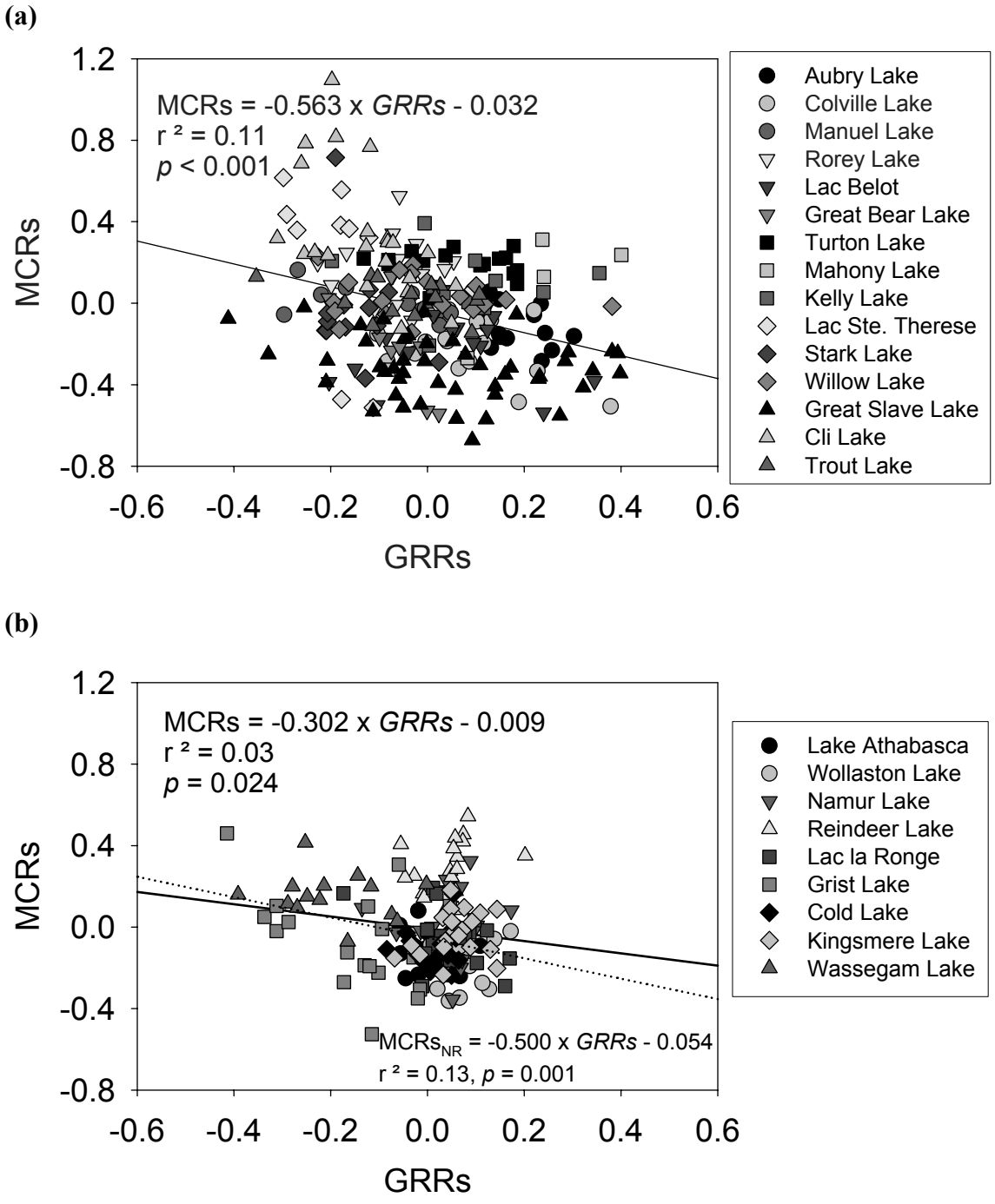


Figure 4.5. Regression of log mercury concentration in lake trout versus fork length residuals (mercury contamination residuals, MCRs) versus negative residuals of age versus fork length (growth rate residuals, GRRs) for lake trout from study lakes in (a) the Northwest Territories, and (b) northern Alberta and Saskatchewan. The dotted regression line represents the same relationship with the lake trout from Reindeer Lake removed, and regression line equation is designated by the subscript 'NR' for 'no Reindeer Lake'.

4.5. Discussion

Previous research has demonstrated that northern lake trout grow slower than more southern populations (Scott and Crossman 2001). The first part of this study confirms that lake trout from the NT study area grew at a slower rate than those sampled from the NAS study lakes (Figure 4.2). While different methods were used to capture the lake trout, which could affect estimates of population size frequency distributions, estimates of growth rates should not be affected. Nor should the time of year in which the fish were caught justify large differences in growth rates, i.e., a 600 mm long fish grows only a few millimetres per year. The greater growth rate variation in NT than NAS lakes is most likely due to the greater number of lakes sampled (15 versus nine), their surface area, and their inaccessibility to anglers.

Growth rate is a reflection of the surrounding environment, including the climate, which in turn affects fish metabolism and lake productivity. Temperature and food sources are primary factors determining the growth potential of an organism (Mason *et al.* 1995, Vøllestad *et al.* 2002. The difference in seasonal lake temperatures between the two study areas likely affects both lake trout metabolism and lake productivity,. On average NT are ice covered for seven to eight months of the year, from October to April. In contrast, NAS lakes are iced covered for approximately six to seven months of the year, from November to April (Table 4.2). The optimal temperature for growth of lake trout is 12.5˚C (Edsall and Cleland 2000), and lake trout grow very little, or not at all in the frozen winter months (Wootton 1992). This study found that the slope of the relationship between log age and fork length is steeper in NT compared to NAS lakes (Figures 4.2a and b) indicating a slower growth rate in the NT lakes. Moreover, as lake trout size increase in length, they are able to feed on larger prey items. Therefore, growth rate is likely more highly correlated to increasing length than weight (Sutton and Ney 2001). Growth rate can decrease with increasing age; this may explain the slower growth rates in the NT lake trout population which tended to be substantially older than the NAS population (Vøllestad *et al.* 2002).

Fishing pressures can also affect lake trout growth rates by contributing to natural mortality, hence reducing population size. Also, larger, older fish are preferentially removed by sport-fishing. This, in turn, reduces competition for food and

enhances growth rates (Verta 1990). In lakes where fishing pressures are light or nonexistent, the population tends to be dominated by older fish; growth rates are lower than in lakes experiencing some fish pressures (Verta 1990).

Fishing pressures are much higher in the NAS study lakes, many of which support both commercial and sport fisheries, than in the NT lakes, which are relatively remote, rarely visited, and with the exception of Great Slave Lake, support no commercial fisheries. The effects of these differences in fishing pressures may be seen in our results. One effect is on the mean age of lake trout in the two study areas. The mean age of the NT lake trout was more than double the mean age of lake trout in the NAS lakes. Low fishing pressures in NT lakes may allow older fish to persist longer than in the NAS lakes, where commercial and sports fisheries, both of which have size (length) limits, continuously remove the larger (older) fish. Second, the average growth rate for lake trout in the NT lakes was much slower than in NAS lakes. While temperature would account for some of this variation, fishing pressures could also be important, i.e., greater competition for food resources in un-fished or lightly fished lakes, particularly from larger adults.

This current study found evidence of a statistically significant inverse relationship between tissue mercury concentration and growth rate in lake trout from lakes in the NT ($p < 0.001$) and NAS ($p = 0.024$) study areas. Other studies have found similar relationships in a variety of species from different geographic areas. Stafford *et al.* (2004) found a negative relationship between mercury concentration and growth rate in lake trout and lake whitefish in Flathead Lake, Montana. In an earlier study Stafford and Haines (2001) found no such relationship in lake trout, but found some evidence in smallmouth bass (*Micropterus dolomieu*); they believed that the lack of a relationship in lake trout was because of the small variation in their growth rates. Doyon *et al.* (1998) determined that dwarf lake whitefish (*Coregonus clupeaformis*), with slow growth rates, bioaccumulated mercury more rapidly than normal individuals in the same reservoir, despite similar diets. Faster growing fish are believed to have lower concentrations of persistent organic contaminants, and other bioaccumulating compounds including mercury, in their tissues because of biodilution (Thomann 1989). Biodilution can be

obscured when growth variation is low, or when there are differences in fish diet or physiology (Stafford and Haines 2001).

Wassegam Lake was the most remote of the NAS lakes with the lightest fishing pressure with access severely limited to hiking. Lake trout were old, slow growing, and had relatively high mercury levels, i.e., they were in the upper left quadrant of the MCR versus GRR regression. Similarly Grist Lake, with limited road access light fishing pressures, has concomitantly slower growing lake trout with higher mercury concentrations. Increased fishing pressures in these lakes and in the NT lakes could result in reduced abundances of fish, particularly older fish, enhanced growth rates and a reduction in mercury concentrations.

A final factor potentially affecting mercury concentrations in fish is local geology. Shilts and Coker (1995) first proposed local geology as an explanatory factor for high mercury levels in lake trout in their study lake. There was no evidence of local geological influences in NT lakes but some evidence for NAS lakes. The relationship between mercury concentration and growth rate is weaker in the NAS than NT study lakes, and this weakness appears to be driven by Reindeer Lake (Figure 4.4 b). Lake trout from Reindeer Lake had the highest mercury concentrations among all the NAS lakes Reindeer Lake is at the end of a corridor running from Lac la Ronge to the northwest, where base metals are sufficiently rich in the bedrock to merit mining activities. Past and present mines in the corridor include a number of gold and copper-zinc mines. There are also a number of undeveloped deposits in the area (Saskatchewan Industry and Resources 2006). The bedrock in these base metal deposits typically is enriched in mercury relative to natural background concentrations (Sexauer Gustin *et al.* 2003). This enriched source of bedrock mercury combined with mercury released from mining activities (leaching from waste rock and tailings ponds) can be significant sources of mercury to the surrounding environment. The relationship between mercury concentration and growth rate was strengthened when Reindeer Lake lake trout were removed from the NAS data set.

4.6. Conclusions

Lake trout from NAS grew at a significantly faster rate that those from NT lakes with the exception of Wassegam Lake. Mercury contamination in lake trout tissue was

weakly and inversely correlated with growth in both geographic areas investigated in this study, i.e., slower growing fish accumulated more mercury in their tissues than did lake trout growing more rapidly. Slower growth rates of fish in the NT than NAS is related to climate i.e., mean annual temperature which affects metabolism and length of growing season which affects productivity and food availability. Fishing pressures also are believed to be important in affecting growth rates and mercury accumulation. While growth rates are weakly correlated with mercury concentration in lake trout, other factors also may be important in affecting differences in mercury concentrations in lake trout from the two study areas. There is the suggestion that local geology may play a role for one of the study lakes, i.e. Lac la Ronge.

5.0 GENERAL DISCUSSION AND CONCLUSIONS

5.1 Synthesis of results

Previous research has shown total mercury (THg) concentrations in lake trout in many lakes in the Northwest Territories (NT) to exceed the Health Canada 0.2 µg Hg/g guideline for frequent consumers of fish (FCFG), and the guideline set by the Canadian food inspection agency (CFIA) for the commercial sale of fish (CSFG) (0.5 µg Hg/g). The main objective of this thesis was to determine why THg tissue concentrations in lake trout from NT lakes were so much higher than in other parts of Canada. I first investigated factors affecting THg concentrations in NT lake trout tissue, developing various hypotheses. Next, in order to more fully explore these hypotheses, the NT data were compared with data collected from a series of lakes in northern Alberta and Saskatchewan (NAS).

I found that THg concentrations in NT lake trout muscle tissue were related to a combination of biological and physical factors. The biological factors included age, fork length, and feeding habits as measured through stable isotope analyses; the physical factors of importance were lake and watershed area. Water chemistry variables such as total phosphorous (TP), nitrogen, dissolved organic carbon (DOC), and pH were not strongly correlated to mercury concentration in lake trout. On a lake-by-lake basis, mercury concentrations were significantly correlated with nitrogen isotopes ($\delta^{15}N$) in all food web species sampled. However, there was no significant relationship between $\delta^{15}N$ values and mercury concentration in lake trout alone, likely due to the narrow range of $\delta^{15}N$ values associated with the lake trout.

Mercury concentrations in NT lake trout increased with fish length and age, with fish tending to reaching 0.5 µg Hg/g at ca. 12 years. Older age has been associated with elevated mercury concentrations in previous studies (MacCrimmon *et al.* 1983; Bodaly *et al.* 1993, Evans and Lockhart 2001; Evans *et al.* 2005), and is seen again here. Lake trout from NT lakes had higher THg concentrations than lake trout of similar fork length (but younger age) from NAS lakes.

Lake surface area was another factor significantly related to THg concentration in NT lake trout. Lakes with small surface areas were associated with higher summer water temperatures because small lakes tend to be shallow and thus reach higher summer maximum temperatures than large, deeper lakes. High water temperatures are more conducive to methylmercury (MeHg) transformations, and ultimately greater THg accumulation within lake trout tissues. Also, small lakes are more likely affected by mercury inputs, bound to organic matter, from the surrounding watershed. In addition to lake surface area, there was also a correlation between THg tissue concentration and the watershed area to lake area ratio. Proportionately larger watershed areas lead to more inputs of mercury and MeHg from the surrounding landscape.

One of the goals of the study was to determine whether it was possible to develop a simple predictive model to assess which lakes are likely to contain lake trout which exceed the 0.2 µg Hg/g and the 0.5 µg Hg/g guidelines. A subset of all the variables investigated for the NT lakes was entered into a multiple regression, with selection based on those variables that were able to account for the most variability in mercury concentration in lake trout tissue. All the variables were first entered into a best subsets regression to determine an accurate and simple model. Latitude, lake trout age and fork length, log lake area and the watershed area to lake surface area ratio emerged as the variables most able to explain the variance in mercury concentration in lake trout tissues. When these variables were entered into a multiple regression the resulting predictive equation is:

$$\log Hg = 0.698 - (0.0156 \times \textit{latitude}) + (0.0031 \times \textit{age}) + (0.000535 \times \text{length}) - (0.245 \times \log \textit{lake area}) + (0.00675 \times \textit{watershed area/lake area ratio}),\ r^2 = 0.73,\ p < 0.001 \quad (2.2)$$

A more simplified version of the equation was created, using all variables except age, which requires specialized procedures to analyze and the death of the fish when otoliths are used (Chapter 2). This equation is well suited as a screening tool which could be used by First Nations and other communities in addition to researchers.

$$\log \textit{Hg} = 0.724 - (0.0174 \times \textit{latitude}) + (0.000588 \times \textit{length}) - (0.2193 \times \log \textit{lake area}) + (0.00197 \times \textit{watershed area/lake area ratio}),\ r^2 = 0.71,\ p < 0.001 \quad (2.3)$$

These two predictive models can be used to coarsely estimate mercury concentrations in the muscle tissue of lake trout from previously unstudied lakes. To be more accurate, a greater number of lakes would need to be studied in more detail. The models also should be tested with data generated in the lakes in the same area but not used in the model construction.

Mercury concentrations in NT lake trout were further investigated through a comparison of mercury biomagnifications rates in NT and NAS lakes. Trophic relationships were studied by measuring carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotopes in aquatic biota; biomagnification rates, measured as the slope of $\delta^{15}N$ versus log THg concentration, were compared between the two study areas. When zooplankton, aquatic invertebrates, forage fish, lake whitefish, and lake trout data were pooled, there was a significant relationship between $\delta^{15}N$ and mercury in NT and NAS lakes. Therefore, tissue mercury concentration in lake trout was highly correlated with trophic position although there was little difference in the mean $\delta^{15}N$ values in lake trout from the two areas. However, there was no correlation between mercury concentrations in lake trout and $\delta^{15}N$ values. Biomagnification rates for NT food webs were lower than those for NAS lakes. Lower biomagnification rates may be a reflection of slower growth and greater age of lake trout in NT than NAS lakes, i.e., mercury concentrations in fish tissue in NT lakes were acquired more slowly, but over an extended lifespan. The y-intercept was higher for NT lakes, suggesting higher concentrations of mercury at the base of the food web. There is a trend showing that all levels of the NT food webs have more negative $\delta^{13}C$ values than their counterparts in the NAS lakes. This is of interest because more negative $\delta^{13}C$ values are associated with higher tissue THg concentrations and a more pelagic diet. This phenomenon was also found in studies by Power *et al.* (2002), Campbell *et al.* (2003) and Kidd *et al.* (2003).

Finally, growth rates of the NT and NAS lake trout were compared to determine growth rate could help explain the higher mercury concentrations NT lake trout. The average fork length of lake trout from both areas was approximately 600 mm. The average age of a 600 mm lake trout was 13.7 years in NT lakes, and 6.7 years in NAS lakes. Lake trout from the NT lakes grew at a slower rate (7.8 mm per year) than those

from NAS lakes (19.2 mm per year). Therefore NT lake trout are slower growing than NAS lake trout.

Tissue THg concentration was significantly correlated to growth rates for lake trout from NT lakes. The equation describing this relationship was

$$MCRs = -0.563 \times GRRs - 0.033 \ (r^2 = 0.11, p < 0.001) \quad (4.7)$$

In the NAS lakes, mercury concentration also was correlated to growth rate, though the relationship was not as strong as in the NT lakes. The equation in the NAS study lakes was:

$$MCRs = -0.302 \times GRRs - 0.009 \ (r^2 = 0.03, p = 0.024) \quad (4.8)$$

Therefore, growth rate is one factor capable of affecting THg concentrations in the tissue of lake trout in the NT lakes and the NAS lakes.

Therefore, high mercury concentrations in muscle tissue of predatory fish in northwestern Canada are correlated to old age, slow growth, feeding habits, and lake surface area and watershed area. Regional differences in these factors contribute to the differences in mean THg concentrations between lake trout from the NT study lakes and the NAS study lakes.

5.2 Significance of research

High mercury concentrations in fish in northern Canada are of great concern to northern residents who rely on fish as a food source. Current reports have found that relative risk and consumption advisories vary widely across the Mackenzie River Basin (MRB), and that the availability of this information to the general public in limited (MRBB 2004). The cost of performing site-specific studies on all the lakes in the MRB is prohibitive, and therefore a method to predict mercury values would be of some use.

Total mercury concentrations in sediments and water bodies are at background levels, and MeHg values are near undetectable (Lockhart *et al.* 2001). Therefore high mercury concentrations in fish tissue are not directly due to an increasingly contaminated environment. Rather, high tissue mercury concentrations are due to factors, including old age, that are conducive to bioaccumulation of mercury. Any increased knowledge, including what is reported in this thesis, concerning the mercury cycle in the aquatic ecosystem will facilitate in the assessment of mercury concentration in species of importance as food sources.

Knowledge of the factors affecting mercury bioaccumulation and biomagnification is important in developing fiscally responsible mercury assessment methods for northern Canada. The predictive models developed in Chapter 2 of this thesis can be used in the mercury assessment of northern lakes. Though they cannot perfectly predict mercury concentrations, they can provide enough information to estimate which lakes might be of concern, and require further study.

Knowledge gained through this thesis implies that future climate change may affect mercury concentrations in aquatic biota through affects on water chemistry, and aquatic biology. Higher air temperatures lead to increased water temperatures; rates of mercury methylation increase with increasing water temperature, potentially leading to an increase in mercury levels in fish. However, climate change also could lead to shorter winter periods, decreasing the amount of time fish are dormant, and grow relatively little. This could increase fish growth rates, potentially decreasing tissue mercury concentration through growth dilution.

5.3 Difficulties encountered

Working in remote northern Canada involves very high travel costs; float planes are required to access most study sites. High costs coupled with fiscal restraint, often imposes restrictions on survey studies, and limits the locations that can be accessed. These difficulties can be overcome to some extent through the collaboration of scientists working in these areas.

The effects of various water chemistry variables on the transformations of mercury in aquatic systems and the bioaccumulation of mercury in lake trout were included in this study, but not extensively as the authors would have liked. This study found very few correlations between water chemistry variables and mercury concentration in fish tissue. However, water chemistry variables are not stable, and undergo moderate change both spatially and temporally. The water chemistry data included in this study was the result of a single collection during the summer months, and only in a subset of the lakes included in the study. The lack of more extensive data is likely what led to the lack of elucidation of firm relationships with mercury tissue concentration. However, funding considerations precluded conducting limnological sampling on lakes in which the original department of fisheries and oceans (DFO) stock

assessment studies were conducted. Future stock assessment studies would benefit from conducting baseline limnological sampling during the assessment, particular water chemistry.

There was high variability among the lakes with respect to lake trout age, length, weight, $\delta^{15}N$ and $\delta^{13}C$ versus mercury relationships (see Chapter 2). Significant relationships were found in some lakes but not others (see Table 2.4), e.g. lake trout from Willow and Mirror lakes displayed no significant relationships. It is still not clear why this is the case. Perhaps genetic differences may account for some of the variation. There is a need to better understand the stock genetic structure within the study lakes.

Lake trout for this study were provided through a variety of collection methods including stock assessments, commercial fisheries and angling. This mixing of collection methods can introduce bias into the results. Angling is an active capture method that requires the fish to be biting, and may select for certain age / size ranges depending on distribution within the lake. Also, anglers may want to return with fish of a certain size, not understanding that a large variation is positive for scientific study. Commercial fisheries may provide 'undesirable' sizes (ie. too large or too small) for the study, while retaining the 'best of the catch' for sale. In addition, there was seasonal variation in the timing of fish capture by gill nets. In general, gill nets were set in the NT lakes in winter months, except Stark and Trout lakes, which were sampled in late summer months. While these differences in methods will affect estimates of the population structure of fish within a lake, they would have a minimal impact on estimates such as growth rates and mercury concentrations at a given size of fish.

5.4 Future directions

As discussed in the previous section, more effort should be put into the investigation of the effects of water chemistry variables on mercury concentration in fish tissues. There are indications that water chemistry affects mercury transformations in more ways than were determined by this study. For example, lake surface area has a strong negative relationship with tissue mercury concentration. How much of this relationship is due to the affect of lake surface area on mean water temperature? Also, watershed to lake surface area ratio was a significant variable that may mask other effects including the DOC of the water. Dissolved organic carbon affects mercury

concentration in many complex ways. When it enters lakes from wetlands it often carries MeHg with it. However, once within a lacustrine environment, it can increase or decrease the rate of transformation of ionic mercury to MeHg (see section 1.4.3.). Future work concerning mercury transformations and bioaccumulation of mercury within the food chain should focus on better understanding of the affects of water chemistry of the mercury cycle in northern Canadian lake systems. Further research into regional and global mercury budgets would help to better hypothesize methods to maintain low mercury concentrations in water and sediment, while lowering concentrations in aquatic organisms.

Further study of fish biology within individual lakes, including growth rates and genetics, may elucidate why we observed no significant relationships between fish length, weight, and age and stable isotope values in some study lakes. Further work such as that of Howland *et al.* (2004) which identified different morphs of lake trout within a population could help explain much of this variability. In addition, our food web work did not include pelagic benthic invertebrates. The sampling of deep-water benthic species may add support to our food web trends.

Another area for further research is climate change. Northern ecosystems are among those most affected by climate change (MRBB 2004). Traditional environmental knowledge tells us that climate in the north is rapidly changing and weather is becoming more variable and difficult to predict. Mean temperatures in the MRB during the winter months have increased 2 to 3°C from 1950 to 1998, and have continued to show a similar (MRBB 2004). Temperature change is important with respect to mercury because higher temperature is related to increased rates of mercury methylation, and ultimately higher mercury concentrations in fish tissue.

Spring melt is beginning earlier and lasting longer, affecting ice break ups and jams in rivers and lakes. Permafrost thickness, particularly in discontinuous zone is lessening. All of these changes will, in all likelihood affect mercury movement and transformations in the environment. The study of mercury cycling has to be a part of the study of future climate change.

In addition to future research continuing to focus on understanding mercury cycles in the northern Canadian aquatic environment, some future work should be devoted to

methods to remove mercury from the ecosystems, or prevent it from accumulating in aquatic organisms. Studies involving the use of intensive fishing to alter food web structure, and ultimately lower mercury concentrations have been ongoing since the early 80s and have been met with varying success (Göthberg 1983; Verta 1990; Masson and Tremblay 2003). Past research has investigated the potential of decreasing mercury in fish by treating lakes with selenium (Paulsson and Lundbergh 1989). Therefore, various methods have been tried with some success in the past. Future work could aim to help alleviate mercury stresses in those lakes in the MRB that are known to house fish populations with high tissue mercury concentrations.

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APPENDIX A

Appendix A includes raw data on all fish caught including age, fork length, weight, body condition, stable carbon ($\delta^{13}C$) isotope ratio, stable nitrogen ($\delta^{15}N$) isotope ratio, and total mercury concentration in muscle tissue.

Table A-1. Lake trout from the Northwest Territory lakes

Lake	Age (years)	Fork Length (mm)	Weight (g)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Hg (µg/g ww)
Aubry	13	700	3700	-25.40	12.15	0.429
Aubry	12	700	4400	-23.84	12.28	0.427
Aubry		750	4160	-22.97	12.17	0.408
Aubry	8	600	2340	-27.54	9.94	0.329
Aubry	9	550	2090	-25.84	11.80	0.325
Aubry		560	2000	-22.78	10.01	0.252
Aubry	8	500	1480	-24.42	10.78	0.197
Aubry	11	640	2620	-24.90	11.57	0.390
Aubry	10	580	1880	-20.91	10.52	0.253
Aubry	12	650	3070	-23.94	11.22	0.372
Aubry	11	650	2930	-24.75	10.63	0.213
Aubry	7	530	1680	-21.52	11.46	0.217
Aubry	8	580	1950	-21.34	11.46	0.283
Aubry	7	520	1750	-27.47	9.48	0.156
Aubry	6	520	1750	-27.89	10.70	0.207
Aubry	9	550	2060	-23.63	11.64	0.219
Aubry	10	620	2840	-26.03	11.16	0.232
Aubry		520	1880	-24.79	10.44	0.129
Aubry	7	550	1580	-21.14	10.58	0.183
Aubry	10	570	2280	-21.72	11.45	0.264
mean	**9**	**592**	**2422**	**-24.14**	**11.07**	**0.270**
st dev	**2**	**70**	**851**	**2.17**	**0.79**	**0.090**
Colville	17	615	2540	-25.48	16.04	0.175
Colville	18	621	3060	-26.22	16.29	0.240
Colville	13	561	2090	-27.66	16.36	0.204
Colville	10	570	1810	-26.12	16.52	0.203
Colville	11	555	1690	-25.58	15.86	0.150
Colville	12	640	2970	-24.92	16.35	0.180
Colville	8	535	1780	-26.66	16.47	0.100
Colville	8	590	1710	-25.95	16.34	0.152
Colville	8	580	1770	-24.48	16.25	0.298
Colville	5	515	1500	-26.41	16.17	0.093
Colville	11	650	2240	-24.03	15.74	0.291
Colville	13	530	1610	-29.46	15.02	0.172
Colville	12	575	2050	-26.52	16.70	0.209
Colville	10	545	1400	-25.48	17.02	0.336

Colville	13	620	2070	-23.67	16.11	0.227
Colville		555	1830	-24.39	15.51	0.207
Colville	10	550	1480	-24.68	16.13	0.254
Colville	12	575	1800	-25.22	16.32	0.274
Colville		565	1640	-25.01	15.86	0.164
Colville	12	580	2060	-24.98	16.61	0.301
mean	**11**	**576**	**1955**	**-25.65**	**16.18**	**0.210**
st dev	**3**	**37**	**455**	**1.33**	**0.44**	**0.070**
Manuel	17	493	1550	-28.71	11.84	0.346
Manuel		465	1310	-28.48	11.04	0.309
Manuel	12	500	1540	-25.71	11.67	0.271
Manuel		480	1470	-29.72	11.54	0.224
Manuel	12	545	1980	-28.29	11.52	0.277
Manuel	11	456	1130	-28.25	11.45	0.261
Manuel		525	1640	-28.47	11.91	0.464
Manuel		500	1150	-28.20	11.37	0.333
Manuel		433	1000	-28.25	10.99	0.256
Manuel	10	440	1030	-28.09	11.39	0.212
Manuel	10	473	1330	-28.96	11.52	0.254
Manuel	13	520	1570	-29.76	11.46	0.334
Manuel	22	510	1370	-27.75	11.76	0.432
Manuel	12	460	1250	-29.04	11.24	0.277
Manuel		490	1340	-27.76	11.98	0.323
Manuel	18	455	1280	-30.21	10.98	0.305
Manuel	22	470	1440	-30.82	11.68	0.248
Manuel		477	1210	-27.44	12.82	0.305
Manuel		511	1630	-26.65	10.56	0.257
mean	**14**	**484**	**1380**	**-28.45**	**11.51**	**0.300**
st dev	**5**	**30**	**240**	**1.20**	**0.48**	**0.060**
Rorey	11	510	1360	-27.18	11.06	0.344
Rorey	12	485	1210	-27.29	11.68	0.423
Rorey	21	582	1730	-27.71	11.39	0.315
Rorey	14	520	1320	-28.66	11.27	0.294
Rorey	13	590	1800	-27.90	11.35	0.294
Rorey	13	520	1580	-30.26	11.05	0.246
Rorey	11	540	1600	-27.98	11.65	0.494
Rorey	14	510	1540	-31.38	10.16	0.406
Rorey	13	550	1720	-28.40	10.94	0.432
Rorey	11	490	940	-30.08	10.68	0.286
Rorey	14	510	1340	-27.35	11.45	0.651
Rorey	14	510	1200	-28.03	11.28	0.481
Rorey	21	580	1820	-27.92	12.00	0.396
Rorey	15	570	1320	-27.24	12.45	0.526
Rorey	21	540	1600	-28.04	11.95	0.485
Rorey	14	530	1480	-28.29	11.22	1.019
Rorey	18	530	1740	-27.46	10.83	0.537
Rorey	10	455	1020	-28.34	11.47	0.407
Rorey	12	484	1330	-27.76	11.63	0.561
Rorey	15	500	1460	-28.33	10.62	0.274

mean	**14**	**525**	**1456**	**-28.28**	**11.31**	**0.440**
st dev	**3**	**36**	**251**	**1.10**	**0.53**	**0.170**
Belot	10	600	3090	-27.17	11.11	0.286
Belot	14	630	2580	-22.35	11.29	0.301
Belot		650	3320	-25.87	11.25	0.344
Belot	6	580	2180	-22.18	10.79	0.134
Belot	18	620	2980	-21.09	9.36	0.107
Belot	15	600	2790	-24.85	11.02	0.192
Belot		620	2720	-22.75	10.10	0.188
Belot	11	640	3150	-26.30	10.72	0.258
Belot	23	630	3150	-23.58	10.78	0.141
Belot	19	680	4030	-23.82	11.11	0.247
Belot	14	700	3800	-25.96	11.32	0.284
Belot	11	620	3190	-25.56	10.80	0.210
Belot		600	2710	-21.95	9.82	0.107
Belot	10	540	1930	-23.35	11.57	0.196
Belot	12	590	2410	-23.78	11.48	0.241
Belot	6	430	950	-21.26	10.19	0.078
Belot	20	620	3260	-21.51	10.70	0.162
Belot		600	3340	-22.54	10.20	0.161
Belot		600	2930	-23.49	10.61	0.101
Belot	11	640	2820	-25.54	11.49	0.338
mean	**13**	**610**	**2867**	**-23.75**	**10.79**	**0.200**
st dev	**5**	**55**	**669**	**1.83**	**0.60**	**0.080**
Great Bear	19	695	4591	-29.60	12.90	0.218
Great Bear	13	600	2320	-25.60	12.40	0.095
Great Bear	15	654	2849	-28.30	12.60	0.105
Great Bear	15	635	2790	-28.60	12.50	0.212
Great Bear	25	865	5077	-26.00	14.90	0.620
Great Bear		912	5864	-25.30	15.20	1.120
Great Bear		690	3334	-28.80	13.10	0.191
Great Bear	22	601	2059	-27.10	13.00	0.351
Great Bear	26	854	5865	-28.40	12.90	0.307
Great Bear	24	863	6116	-28.20	13.40	0.280
mean	**20**	**737**	**4086**	**-27.58**	**13.29**	**0.350**
st dev	**5**	**123**	**1588**	**1.49**	**0.97**	**0.310**
Turton	10	570	1780	-32.01	12.07	0.497
Turton	18	580	2370	-32.27	12.16	0.535
Turton	16	580	2320	-31.46	11.73	0.526
Turton	9	570	1780	-32.50	12.15	0.534
Turton	11	540	1720	-31.94	12.37	0.581
Turton	16	580	2240	-32.72	11.97	0.503
Turton	14	600	2540	-31.48	12.39	0.535
Turton	8	530	1680	-31.61	12.25	0.378
Turton	13	520	1040	-29.93	12.01	0.540
Turton		560	2300	-31.62	11.32	0.539
Turton		560	1890	-31.43	11.86	0.558
Turton	10	560	1780	-30.06	12.34	0.484

Turton	8	520	1440	-30.83	12.34	0.573
Turton	13	620	2470	-32.44	11.35	0.582
Turton	8	530	1460	-32.20	11.74	0.441
Turton		600	2500	-32.27	12.34	0.544
Turton	9	550	1580	-30.99	12.18	0.515
Turton	11	470	1150	-30.96	11.78	0.299
Turton	8	520	1450	-30.44	12.49	0.420
Turton		610	2520	-31.16	11.97	0.566
mean	**11**	**559**	**1901**	**-31.52**	**12.04**	**0.510**
st dev	**3**	**37**	**475**	**0.80**	**0.33**	**0.070**
Mahony		720	4480	-26.15	11.27	0.521
Mahony	12	635	3110	-27.84	10.36	0.186
Mahony		500	1430	-25.18	10.71	0.144
Mahony	15	540	1960	-27.86	9.88	0.145
Mahony	13	905	8730	-24.63	11.32	0.989
Mahony		860	7650	-25.63	11.10	0.811
Mahony	10	975	10000	-25.34	11.40	0.907
Mahony	12	860	8020	-24.72	11.28	0.615
Mahony		540	1810	-27.12	9.06	0.201
Mahony		545	2020	-26.26	10.70	0.176
Mahony		540	1810	-26.99	9.13	0.173
Mahony		535	1840	-25.96	10.69	0.167
Mahony		560	2010	-24.84	10.75	0.167
Mahony		530	1590	-25.96	10.80	0.174
Mahony		550	1730	-26.44	9.37	0.157
Mahony		625	3120	-26.64	11.00	0.335
Mahony		640	3110	-26.46	10.26	0.226
Mahony		695	3750	-25.72	10.56	0.179
Mahony		805	5160	-25.21	11.91	0.659
Mahony		870	10000	-26.20	10.88	0.404
mean	**12**	**672**	**4167**	**-26.06**	**10.62**	**0.370**
st dev	**2**	**153**	**3001**	**0.94**	**0.76**	**0.280**
Kelly		585	1910	-31.89	12.46	0.593
Kelly	14	660	1800	-29.13	13.18	0.400
Kelly	7	525	1730	-31.46	12.52	0.342
Kelly		590	2420	-28.88	12.62	0.369
Kelly	7	690	3650	-29.35	13.28	0.520
Kelly	21	855	6540	-30.33	12.47	1.120
Kelly	11	480	1310	-31.82	12.41	0.343
Kelly	15	660	2800	-31.72	12.01	0.221
Kelly		620	2910	-30.41	13.11	0.523
Kelly		570	2040	-32.37	12.29	0.363
Kelly	11	630	2680			0.326
Kelly		600	4220	-30.18	12.39	0.731
Kelly	10	605	2600	-30.25	12.20	0.429
Kelly		590	2300	-30.11	11.98	0.490
Kelly		550	1870	-29.42	12.42	0.521
Kelly	10	545	2010	-30.26	12.26	0.501
Kelly		510	1730	-29.91	11.74	0.507

Kelly	19	520	2390	-30.13	12.47	0.484
Kelly		545	1900	-30.12	12.82	0.299
Kelly	10	525	1630	-30.03	12.32	0.306
mean	**12**	**593**	**2522**	**-30.41**	**12.47**	**0.470**
st dev	**5**	**83**	**1178**	**0.99**	**0.40**	**0.190**
Mirror		474	1260	-29.25	8.64	0.530
Mirror		528	1770	-28.38	9.74	0.974
Mirror		496	1430	-33.18	8.24	0.589
Mirror		484	1320	-28.68	9.88	0.679
Mirror		442	1010	-30.41	9.82	0.532
Mirror		509	1500	-30.26	9.61	0.625
Mirror		432	1020	-32.20	9.80	0.508
Mirror		450	1010	-28.66	9.89	0.540
Mirror		529	1670	-28.94	8.65	1.085
Mirror		480	1290	-29.32	9.74	0.517
Mirror		452	1140	-28.22	9.85	0.800
Mirror		570	1830	-28.31	9.94	0.481
Mirror		498	1470	-28.78	10.39	0.588
Mirror		431	940	-29.11	9.41	1.651
Mirror		421	940	-29.69	10.08	0.952
Mirror		425	910	-29.55	10.66	0.572
Mirror		438	1000	-31.28	9.53	0.474
Mirror		439	1070	-33.18	9.44	0.481
Mirror		454	1190	-29.50	9.85	0.460
Mirror		425	1010	-29.61	10.04	0.598
mean		**469**	**1239**	**-29.83**	**9.66**	**0.680**
st dev		**42**	**288**	**1.52**	**0.58**	**0.290**
Ste. Thérèse	24	690	2313	-28.10	14.10	0.893
Ste. Thérèse	20	580	2578	-27.70	12.10	0.109
Ste. Thérèse	15	790	4403	-28.60	13.10	0.489
Ste. Thérèse	30	700	2400	-27.80	14.00	0.856
Ste. Thérèse	26	765	3506	-29.40	13.70	0.943
Ste. Thérèse	31	690	2689	-28.10	14.00	1.010
Ste. Thérèse	22	720	2765	-28.10	13.30	0.806
Ste. Thérèse	25	718	3574	-28.70	14.60	1.380
Ste. Thérèse	24	786	4877	-28.50	12.70	0.128
Ste. Thérèse		650	3895	-26.80	12.50	0.315
Ste. Thérèse	32	700	2396	-28.10	14.40	1.550
mean	**25**	**708**	**3218**	**-28.17**	**13.49**	**0.770**
st dev	**5**	**61**	**888**	**0.67**	**0.82**	**0.470**
Stark	17	618	2300	-22.60	12.22	0.383
Stark	19	630	2560	-25.55	12.93	0.327
Stark		640	2760	-28.70	11.83	0.154
Stark	14	646	2800	-27.02	11.89	0.180
Stark	24	646	3040	-23.48	12.94	0.258
Stark	20	650	4120	-29.40	11.83	0.151
Stark		655	2720	-30.38	12.84	0.346
Stark		685	3200	-27.29	12.47	0.395

Stark	26	698	3720	-27.44	12.18	0.274
Stark	26	701	3140	-28.36	12.21	0.305
Stark	21	715	3960	-27.20	12.51	0.296
Stark		715	2540	-24.50	13.08	1.112
Stark		716	4400	-26.73	11.88	0.313
Stark	26	725	4620	-23.96	14.99	2.005
Stark	25	725	3660	-27.19	12.00	0.379
Stark		745	4900	-26.29	12.55	0.359
Stark		764	5560	-31.84	11.80	0.251
Stark	30	790	5780	-24.85	12.78	0.374
Stark	25	800	5280	-24.50	13.18	0.494
Stark	28	800	6740	-24.41	12.35	0.324
Stark		804	6120	-21.86	12.30	0.635
Stark		835	7340	-27.05	12.31	0.579
Stark		857	9780	-25.22	13.80	0.990
Stark		867	8380	-25.02	12.70	0.313
mean	**23**	**726**	**4559**	**-26.29**	**12.57**	**0.470**
st dev	**5**	**74**	**1990**	**2.42**	**0.72**	**0.400**
Willow		598	2340	-29.70	12.80	0.344
Willow		635	2960	-26.10	11.50	0.286
Willow	14	570	1530	-27.40	13.80	0.493
Willow	20	700	3710	-26.10	12.50	0.383
Willow		560	1890	-27.70	12.40	0.383
Willow		550	1920	-28.30	14.10	0.405
Willow	11	560	2110	-28.10	13.00	0.342
Willow	22	570	1900	-27.00	13.40	0.539
Willow		504	1440	-28.10	13.20	0.161
Willow		575	2120	-28.10	13.50	0.463
Willow	13	519	1520	-28.00	13.50	0.421
Willow	8	810	6580	-26.60	12.40	0.414
Willow		803	6230	-27.40	12.30	0.360
Willow	15	790	5920	-25.60	12.30	0.389
Willow		584	2480	-28.00	13.60	0.390
Willow		580	2230	-27.90	13.50	0.445
Willow	19	845	10500	-25.50	12.10	0.441
Willow	20	712	4260	-25.70	12.80	0.335
Willow	21	588	1940	-28.90	13.00	0.329
Willow		527	1780	-28.70	13.00	0.453
Willow		577	2430	-30.00	12.70	0.328
Willow	19	755	8460	-27.10	12.30	0.288
Willow	22	633	3330	-27.90	13.00	0.256
Willow	10	548	2400	-28.00	14.10	0.379
Willow	10	572	1710	-26.90	13.00	0.316
Willow	9	570	1940	-28.20	13.20	0.332
Willow	13	566	2140	-28.60	13.10	0.405
Willow	21	591	2550	-28.20	12.90	0.300
Willow	15	573	2090	-28.30	13.90	0.466
Willow		562	1760	-28.10	13.60	0.506
Willow		575	2150	-28.20	13.90	0.435
Willow	18	536	1840	-27.70	13.50	0.387

mean	**16**	**614**	**3068**	**-27.70**	**13.07**	**0.380**
st dev	**5**	**93**	**2160**	**1.08**	**0.63**	**0.080**
Great Slave	5	543	1740	-28.60	12.90	0.140
Great Slave	6	478	1325	-28.50	13.30	0.080
Great Slave	6	649	3640	-29.60	13.20	0.200
Great Slave	6	547	1860	-28.60	12.20	0.120
Great Slave	6	632	3100	-29.50	12.40	0.200
Great Slave	6	575	2140	-28.90	12.40	0.150
Great Slave	7	589	2500	-30.70	12.00	0.170
Great Slave	7	599	2800	-30.30	12.10	0.190
Great Slave	7	514	2000	-29.50	12.20	0.130
Great Slave	8	465	1250	-25.50	12.10	0.100
Great Slave	8	593	2550	-29.20	12.40	0.140
Great Slave	8	495	1590	-28.90	12.70	0.130
Great Slave	9	512	1660	-25.30	11.70	0.080
Great Slave	10	436	1060	-22.10	11.50	0.110
Great Slave	10	649	3000	-29.40	12.80	0.170
Great Slave	10	604	2360	-28.50	12.70	0.130
Great Slave	11	595	2710	-25.70	11.90	0.070
Great Slave	11	538	2120	-28.10	12.40	0.200
Great Slave	11	575	2240	-29.00	12.10	0.180
Great Slave	12	602	3150	-27.90	12.60	0.090
Great Slave	12	515	1495	-27.60	12.20	0.190
Great Slave	12	672	4590	-28.20	13.00	0.180
Great Slave	13	427	840	-25.90	12.40	0.130
Great Slave	13	474	1400	-27.30	11.60	0.100
Great Slave	14	695	3590	-27.90	12.30	0.140
Great Slave	14	541	2100	-27.80	11.60	0.140
Great Slave	14	602	2640	-27.60	12.50	0.300
Great Slave	14	875	10390	-28.20	13.80	0.410
Great Slave	15	584	2300	-27.40	12.40	0.100
Great Slave	15	556	2400	-27.10	11.90	0.150
Great Slave	15	635	3080	-29.30	11.60	0.110
Great Slave	15	598	2350	-29.40	12.40	0.230
Great Slave	15	529	2000	-28.20	11.90	0.140
Great Slave	15	584	3280	-28.70	13.50	0.170
Great Slave	16	535	1790	-23.10	11.40	0.090
Great Slave	17	649	3700	-27.90	12.10	0.150
Great Slave	18	639	3250	-27.70	13.40	0.290
Great Slave	18	699	5450	-30.60	14.10	0.250
Great Slave	18	760	6040	-31.20	13.60	0.210
Great Slave	19	605	2650	-28.50	12.20	0.260
Great Slave	20	653	2630	-28.00	12.50	0.230
Great Slave	21	565	2000	-29.50	12.20	0.130
Great Slave	21	500	1540	-23.50	11.30	0.280
Great Slave	25	676	3380	-27.90	11.30	0.190
Great Slave	28	573	2200	-29.00	12.40	0.180
Great Slave	34	575	2000	-31.00	12.90	0.270
mean	**13**	**585**	**2693**	**-28.09**	**12.39**	**0.170**
st dev	**6**	**83**	**1558**	**1.91**	**0.66**	**0.070**

Cli	31	835	7760	-25.10	13.80	2.896
Cli		450	880	-24.90	10.60	0.548
Cli	13	390	650	-26.00	12.70	0.575
Cli	11	405	700	-28.40	12.40	0.295
Cli	12	399	620	-26.40	11.90	0.535
Cli	18	405	680	-28.00	11.50	0.455
Cli	28	670	3620	-24.80	12.80	1.752
Cli	30	725	4160	-25.10	13.40	2.355
Cli	25	690	3050	-23.90	13.50	4.612
Cli	9	340	340	-26.30	11.30	0.423
Cli	13	450	1030	-26.90	10.90	0.549
Cli		500	1360	-26.80	10.80	0.391
Cli	14	435	980	-26.70	11.30	0.327
Cli	14	432	870	-28.00	10.40	0.511
Cli	8	350	460	-27.00	11.30	0.296
Cli	27	850	5400	-24.80	13.40	2.636
Cli		500	1160	-27.30	10.00	0.569
Cli	19	470	1030	-27.80	9.80	0.503
Cli	12	440	750	-26.70	11.10	0.204
Cli	9	410	710	-27.50	11.90	0.209
Cli	12	465	1140	-26.80	10.50	0.372
Cli	18	475	1000	-27.10	11.80	0.488
Cli	14	490	1090	-27.10	12.30	0.463
Cli	13	465	940	-27.50	12.80	0.556
Cli	22	450	900	-26.20	9.10	0.574
mean	**17**	**500**	**1651**	**-26.53**	**11.65**	**0.920**
st dev	**7**	**140**	**1792**	**1.18**	**1.25**	**1.080**
Little Doctor		561	1670	-30.20	12.50	0.488
Little Doctor		512	1260	-30.80	12.80	0.444
Little Doctor		586	1820	-30.20	12.40	0.445
Little Doctor		532	1610	-30.20	12.70	0.377
Little Doctor		582	1900	-27.70	12.40	0.400
Little Doctor		539	1400	-29.30	11.60	0.408
Little Doctor		607	2120	-29.70	12.40	0.451
Little Doctor		458	890	-30.20	13.00	0.198
Little Doctor		556	1770	-30.80	11.70	0.376
Little Doctor		540	1590	-30.00	11.80	0.342
mean		**547**	**1603**	**-29.91**	**12.34**	**0.390**
st dev		**42**	**350**	**0.91**	**0.50**	**0.080**
Trout	18	602	2950	-28.16	12.98	0.453
Trout	18	731	4930	-26.54	12.13	0.338
Trout	21	620	1960	-28.34	12.64	0.340
Trout	32	620	2040	-26.53	10.91	0.458
Trout	16	648	2510	-28.76	12.36	0.391
Trout	14	650	3030	-26.57	12.03	0.430
Trout	18	621	2040	-28.53	11.87	0.455
Trout	14		2730	-27.25	12.32	0.383
Trout	12	663	2370	-27.22	11.63	0.369

Trout	17	629	2420	-27.49	12.80	0.312
Trout	13	590	2130	-28.33	12.19	0.349
Trout	20	672	2680	-26.11	12.04	0.280
Trout	12	648	2550	-26.58	11.01	0.252
Trout		700	2530	-25.79	12.04	0.512
Trout	12	670	2490	-28.20	12.11	0.398
Trout	14	624	2200	-28.07	12.70	0.421
Trout		720	3790	-26.26	12.60	0.446
mean	**17**	**651**	**2668**	**-27.34**	**12.14**	**0.390**
st dev	**5**	**41**	**734**	**0.97**	**0.57**	**0.070**

Table A-2. Lake trout from the northern Alberta and Saskatchewan lakes.

Lake	Age (years)	Fork Length (mm)	Weight (g)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	Hg (μg/g ww)
Athabasca	7	713	4343.9	-26.8	12.7	0.293
Athabasca	10	796	4861.3	-26.1	12.5	0.392
Athabasca	7	723	4196.3	-29.3	11.4	0.192
Athabasca	7	736	3993.3	-26.7	12.4	0.310
Athabasca	7	635	2907.7	-28.9	12.6	0.384
Athabasca	8	699	3200.7	-27.0	12.1	0.364
Athabasca	7	815	5832.2	-	-	0.325
Athabasca	7	704	3323.3	-	-	0.180
Athabasca	9	741	4334.7	-	-	0.188
Athabasca	7	746	4250.3	-	-	0.211
Athabasca	9	723	4436.8	-	-	0.238
Athabasca	10	861	7795.2	-	-	0.262
Athabasca	7	630	3326.2	-	-	0.201
Athabasca	10	852	8619.6	-	-	0.350
Athabasca	7	746	4586.8	-25.7	11.6	0.195
Athabasca	7	746	4676.0	-30.7	11.4	0.326
Athabasca	8	750	4654.5	-26.1	12.3	0.287
Athabasca	7	741	3966.5	-26.8	10.8	0.239
Athabasca	7	750	5249.8	-26.2	11.7	0.223
Athabasca	8	736	3907.7	-25.7	12.9	0.307
mean	**8**	**742**	**4623.1**	**-27.2**	**12.0**	**0.270**
st. dev.	**1**	**58**	**1416.9**	**1.6**	**0.6**	**0.070**
Wollaston	5	477	1290.3	-26.6	13.0	0.146
Wollaston	4	459	1386.2	-26.4	12.4	0.147
Wollaston	5	490	1544.9	-25.9	12.5	0.179
Wollaston	5	505	1687.5	-26.9	13.0	0.179
Wollaston	5	536	1794.1	-26.9	12.7	0.131
Wollaston	4	441	1152.5	-26.3	12.9	0.080
Wollaston	6	587	2550.5	-26.7	13.0	0.151
Wollaston	4	418	1289.0	-26.4	13.3	0.081
Wollaston	5	492	1568.3	-	-	0.160
Wollaston	5	501	1602.4	-27.4	13.0	0.173
Wollaston	5	501	1591.5	-27.0	12.6	0.163
Wollaston	6	533	2045.2	-27.1	12.2	0.165
Wollaston	6	606	2430.5	-26.6	12.9	0.218
Wollaston	5	473	1214.2	-27.5	13.1	0.146
Wollaston	5	464	1299.4	-25.9	13.2	0.074
Wollaston	5	424	1017.6	-26.6	11.4	0.077
Wollaston	4	515	1816.8	-24.6	12.7	0.184
Wollaston	5	529	2070.8	-28.1	12.8	0.158
Wollaston	5	501	1464.8	-27.8	13.2	0.150
Wollaston	5	501	1636.7	-26.0	12.7	0.084
mean	**5**	**498**	**1622.7**	**-26.7**	**12.8**	**0.140**
st. dev.	**1**	**47**	**406.3**	**0.8**	**0.4**	**0.040**

Namur	7	527	3114.0	-22.8	13.8	0.187
Namur	5	560	2042.9	-23.1	13.3	0.219
Namur	4	517	1518.9	-23.5	13.2	0.234
Namur	6	670	4342.9	-22.8	13.8	0.594
Namur	6	499	1226.0	-23.1	13.4	0.178
Namur	7	641	2882.0	-24.3	13.1	0.388
Namur	7	699	2954.8	-22.4	13.2	0.517
Namur	7	577	2216.7	-23.1	13.2	0.179
Namur	6	637	3264.3	-23.0	13.1	0.409
Namur	5	475	1055.6	-22.5	13.7	0.180
Namur	6	609	2583.8	-22.3	13.0	0.107
Namur	5	515	1404.9	-22.5	13.5	0.191
Namur	10	667	3634.3	-23.6	13.4	0.347
Namur	6	540	1705.2	-22.2	13.2	0.206
Namur	7	688	3812.1	-23.9	14.1	0.446
Namur	5	505	1509.2	-23.0	12.7	0.119
Namur	6	603	2410.9	-24.3	13.6	0.282
Namur	5	510	1452.3	-23.2	13.5	0.132
mean	**6**	**580**	**2396.2**	**-23.1**	**13.4**	**0.270**
st. dev.	**1**	**73**	**982.5**	**0.6**	**0.3**	**0.140**
Reindeer	6	507	1647.1	-28.5	11.7	0.277
Reindeer	5	487	1295.7	-27.1	11.2	0.271
Reindeer	5	468	1052.7	-27.9	11.7	0.325
Reindeer	5	493	1381.2	-27.4	12.0	0.405
Reindeer	5	514	1567.4	-26.8	12.0	0.550
Reindeer	4	565	2118.6	-26.4	11.6	0.490
Reindeer	7	589	2229.1	-26.9	12.0	0.413
Reindeer	5	473	1250.1	-26.9	11.5	0.307
Reindeer	7	556	1743.5	-26.5	11.8	0.373
Reindeer	5	512	1788.1	-27.8	12.2	0.504
Reindeer	6	618	3304.3	-26.1	12.7	0.683
Reindeer	6	575	2409.1	-27.0	11.8	0.320
Reindeer	6	612	2435.4	-27.1	12.1	0.596
Reindeer	6	584	2319.0	-27.4	11.5	0.277
Reindeer	5	495	1431.3	-27.8	11.8	0.223
Reindeer	5	530	1804.3	-27.3	11.9	0.701
Reindeer	5	492	1330.4	-27.3	11.7	0.352
Reindeer	6	519	1541.0	-27.9	12.0	0.286
Reindeer	6	510	1736.5	-28.1	11.5	0.266
Reindeer	7	542	1955.4	-27.4	11.5	0.527
mean	**6**	**532**	**1817.0**	**-27.3**	**11.8**	**0.410**
st. dev.	**1**	**45**	**533.6**	**0.6**	**0.3**	**0.140**
la Ronge	6	727	4302.2	-26.4	14.7	0.312
la Ronge	8	778	6530.4	-27.7	15.8	0.250
la Ronge	8	750	3999.5	-26.2	14.7	0.534
la Ronge	7	616	2579.6	-26.5	13.9	0.127
la Ronge	3	304	271.2	-25.3	12.2	0.081
la Ronge	7	653	2448.8	-25.7	14.1	0.226
la Ronge	8	441	1499.4	-25.9	14.1	0.236

la Ronge	6	667	3732.9	-28.1	13.0	0.264
la Ronge	4	358	507.4	-26.0	12.9	0.121
la Ronge	9	815	6114.0	-26.0	14.9	0.390
la Ronge	3	288	247.6	-25.5	11.9	0.057
la Ronge	7	681	3715.6	-27.1	14.0	0.262
la Ronge	4	399	778.2	-26.2	13.5	0.097
la Ronge	5	408	728.2	-25.5	13.5	0.123
la Ronge	9	847	9540.0	-25.8	14.9	0.633
la Ronge	7	796	7884.0	-27.9	14.3	0.414
la Ronge	8	723	4533.3	-28.2	13.7	0.238
la Ronge	5	501	1292.2	-25.7	13.9	0.165
la Ronge	5	468	1060.3	-26.2	14.3	0.118
mean	**6**	**591**	**3250.8**	**-26.4**	**13.9**	**0.240**
st. dev.	**2**	**185**	**2732.8**	**0.9**	**1.0**	**0.160**
Grist	5	364	503.5	-27.8	11.2	0.066
Grist	8	454	1048.3	-28.4	11.9	0.125
Grist	14	616	2639.7	-28.3	12.2	0.236
Grist	7	465	1035.2	-28.4	11.8	0.102
Grist	14	574	2562.3	-28.3	12.6	0.250
Grist	8	682	4539.9	-28.1	12.5	0.206
Grist	9	611	2948.8	-28.0	12.1	0.308
Grist	8	574	2313.0	-28.4	11.6	0.218
Grist	13	562	2310.0	-30.9	12.1	0.275
Grist	8	514	1464.9	-28.3	12.1	0.125
Grist	14	657	3966.6	-29.5	12.1	0.289
Grist	19	666	4181.4	-28.6	12.8	0.805
Grist	9	717	6108.7	-26.8	12.9	0.640
Grist	8	539	1963.0	-29.1	12.2	0.061
Grist	6	489	1508.8	-28.0	11.6	0.081
Grist	9	528	1551.4	-28.5	11.7	0.107
Grist	8	531	1796.4	-28.4	11.9	0.129
Grist	7	711	4622.6	-26.6	12.3	0.395
mean	**10**	**570**	**2614.7**	**-28.4**	**12.1**	**0.250**
st. dev.	**4**	**96**	**1508.9**	**0.9**	**0.4**	**0.200**
Cold	7	644	3953.9	-27.7	12.5	0.160
Cold	7	723	5285.1	-27.2	13.2	0.467
Cold	8	778	5672.2	-25.5	13.1	0.303
Cold	6	630	3780.8	-27.0	12.6	0.176
Cold	7	732	4627.6	-25.9	12.8	0.290
Cold	7	704	4792.6	-26.2	12.3	0.226
Cold	5	473	1504.3	-25.3	12.8	0.101
Cold	7	676	4899.6	-27.6	12.5	0.175
Cold	7	621	3377.5	-25.3	13.1	0.169
Cold	8	760	6373.3	-25.4	13.1	0.245
Cold	8	658	4874.8	-26.0	13.3	0.256
Cold	8	671	5223.4	-26.0	13.1	0.242
Cold	8	723	5964.7	-27.5	12.4	0.215
Cold	7	704	5742.1	-25.9	12.9	0.223
Cold	8	695	4746.0	-26.5	13.0	0.222

Cold	9	676	4613.1	-26.4	13.8	0.222
mean	**7**	**679**	**4714.4**	**-26.3**	**12.9**	**0.230**
st. dev.	**1**	**70**	**1169.1**	**0.8**	**0.4**	**0.080**
Kingsmere	6	704	3636.3	-23.8	14.3	0.359
Kingsmere	5	630	2953.7	-24.4	13.6	0.313
Kingsmere	5	538	1883.1	-24.5	14.3	0.163
Kingsmere	6	616	2701.2	-25.5	13.7	0.235
Kingsmere	7	579	1968.8	-25.3	13.0	0.183
Kingsmere	4	468	1104.5	-25.8	13.8	0.108
Kingsmere	6	649	3120.4	-23.9	14.7	0.334
Kingsmere	6	602	2125.3	-24.5	14.6	0.363
Kingsmere	6	496	1461.8	-25.4	15.0	0.135
Kingsmere	6	602	2310.0	-24.8	14.2	0.293
Kingsmere	6	676	3418.5	-24.2	14.4	0.304
Kingsmere	6	575	2197.6	-24.7	14.2	0.249
Kingsmere	7	718	3904.6	-24.0	13.5	0.336
Kingsmere	6	642	2985.5	-25.1	13.4	0.254
Kingsmere	6	584	2213.2	-24.9	14.5	0.195
Kingsmere	6	630	3137.6	-24.7	13.6	0.232
Kingsmere	7	521	1714.0	-26.3	14.8	0.138
Kingsmere	5	607	2520.8	-25.9	13.9	0.188
Kingsmere	6	577	2078.7	-23.7	14.3	0.131
Kingsmere	7	690	3201.5	-24.8	14.4	0.236
mean	**6**	**605**	**2531.9**	**-24.8**	**14.1**	**0.240**
st. dev	**1**	**67**	**750.0**	**0.7**	**0.5**	**0.080**
Wassegam	16	580	2180.0	-27.9	12.7	0.327
Wassegam	9	541	1800.0	-28.0	12.6	0.175
Wassegam		555	1820.0	-28.1	12.7	0.447
Wassegam		562	1870.0	-27.0	13.2	0.236
Wassegam		584	1790.0	-26.4	12.3	0.301
Wassegam	11	496	1330.0	-28.3	12.9	0.292
Wassegam	11	547	1670.0	-28.5	12.5	0.295
Wassegam	8	491	1200.0	-28.3	12.4	0.326
Wassegam	6	519	1280.0	-28.4	13.1	0.317
Wassegam	11	590	2050.0	-28.6	13.1	0.316
Wassegam	7	511	1410.0	-28.1	12.6	0.221
Wassegam	12	543	1560.0	-28.5	12.9	0.270
Wassegam		544	1460.0	-28.0	13.2	0.420
Wassegam	10	536	1670.0	-27.4	12.2	0.325
Wassegam		545	1320.0	-27.5	12.4	0.445
Wassegam	11	540	1410.0	-28.3	13.0	0.534
Wassegam		570	1920.0	-27.7	12.8	0.467
Wassegam	7	530	1530.0	-28.7	12.5	0.214
Wassegam	8	536	2050.0	-28.4	11.9	0.322
Wassegam	11	512	1420.0	-27.5	12.5	0.239
mean	**10**	**542**	**1637.0**	**-28.0**	**12.7**	**0.320**
st. dev	**3**	**27**	**287.3**	**0.6**	**0.3**	**0.090**

APPENDIX B

Graphs showing regressions for variables significantly correlated with mercury concentration in muscle tissue of lake trout from a collection of 17 lakes in the Northwest Territories, Canada.